Assessment of PCDD/Fs, dioxin-like PCBs and PBDEs in Mediterranean striped dolphins

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ABSTRACT

Bio-accumulation of high levels of persistent organic pollutants represent a serious conservation concern for Mediterranean marine odontocetes. In this study, blubber samples from 10 striped dolphins (*Stenella coeruleoalba*) stranded along the Italian coasts during 2015–2016 were analyzed. All specimens showed dl-PCBs > PBDEs ≫ PCDD/Fs. Median concentrations were 1820 ng/g l.w., 456 ng/g l.w. and 23.9 pg/g l.w., respectively. dl-PCBs accounted for 93.3% of total TEQs. PBDE concentrations suggest that the Mediterranean basin may be considered a hotspot for organobromine compounds. OCDD did not represent the greatest contributor to PCDD/Fs profile, most likely due to a change in dioxin environmental sources in the last two-three decades. Despite international regulations, the present study emphasized that POP exposure levels in Mediterranean striped dolphins have not declined significantly in recent years. Toxicological and risk assessment studies on this sentinel species may provide an early indication of potential adverse health effects on Mediterranean ecosystems.

1. Introduction

The Mediterranean Sea is a highly biologically diverse marine ecosystem featuring an exceptional concentration of endemic species (Coll et al., 2010). However, its endemism and high species richness are reported to be among the most threatened at a global scale, due to multiple pressures of human-related activities acting simultaneously and chronically in this area (Halpern et al., 2008; UNEP/MAP, 2012). Chemical and noise pollution due to maritime traffic, habitat loss and degradation caused by growing tourism and coastal urbanization, food depletion (due to overfishing) and bycatch are the main threats affecting marine mammal populations' stability and biodiversity (Marsili et al., 2018). Cetaceans play a crucial ecological role as predators in marine food webs (Pace et al., 2015). In the case of odontocetes, their trophic position, high body-fat percentage, long life expectancies and reduced metabolization capacity compared to pinnipeds and terrestrial species (Hoydal et al., 2018; Reijnders et al., 2018; Sonne et al., 2018) make them more prone to be adversely affected by persistent organic pollutants (POPs) (Casalone et al., 2014; Grattarola et al., 2019; Law, 2014; Mazzariol et al., 2016). As a consequence, their usefulness as sentinel species for marine pollution has been demonstrated, providing an early warning on the cumulative and synergistic effects of different environmental stressors on ecosystems and human health (Bossart, 2011).

POPs such as polychlorinated biphenyls (PCBs), polychlorinated dibenzo-*p*-dioxins and furans (PCDD/Fs) and polybrominated diphenyl ethers (PBDEs) are lipophilic organic compounds resistant to environmental degradation (UNEP/MAP, 2012). PCDD/Fs and PCBs are mainly introduced into the environment as by-products of industrial and combustion processes (She et al., 2016). On the other hand, PBDEs have been produced since the 1970s at three different degrees of bromination (PentaBDE, OctaBDE, DecaBDE commercial mixtures) and extensively used as additive flame retardant compounds (Alaee et al., 2003). Because of mounting evidences on adverse health effects, PCB production and use was banned by most developed countries in the late 1970s. Subsequently, PCB production, use and unintentional release of PCDD/Fs were globally regulated in 2001 through the Stockholm Convention (Stockholm Convention, 2001). Penta- and Octa-BDE and Deca-BDE mixtures were banned in the European Union (EU) in 2004 (La Guardia et al., 2006), and 2008 (European Union, 2008) and listed under Annex A of the Stockholm Convention in 2009 (Stockholm Convention, 2009a, 2009b) and 2017 (Stockholm Convention, 2017), respectively.

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A variety of adverse health effects in humans and wildlife have been reported for POPs (Alonso et al., 2014; Desforges et al., 2018). Toxic responses to persistent organic pollutants includes carcinogenicity (IARC Working Group on the Evaluation of Carcinogenic Risks to Humans, 2016; IARC Working Group on the Evaluation of Carcinogenic Risks to Humans, 2012), immunotoxicity (Centelleghe et al., 2019; De Guise et al., 1995; Desforges et al., 2018; Hammond et al., 2005; Jepson et al., 2005; Marsili et al., 2019), endocrine disruption (Jiménez, 1997; Linares et al., 2015; Mikula and Svobodová, 2006; Schwacke et al., 2012) and adverse effects on reproduction, offspring survivorship rates (Costa et al., 2014; Roos et al., 2012), and population growth (Hall et al., 2018; Jepson et al., 2016). In this context, exposure to immunotoxic PCBs has been suspected to play a significant role in the morbillivirus epizootic outbreaks in striped dolphins (*Stenella coeruleoalba*) in Mediterranean waters during 1990/92 and the first quarter of 2013 (Aguilar and Borrell, 1994; Casalone et al., 2014; Marsili et al., 1997). High levels of organochlorine compounds have been reported in a Mediterranean fin whale (*Balaenoptera physalus*), in association with a simultaneous *Dolphin Morbillivirus* and *Toxoplasma gondii* coinfection (Mazzariol et al., 2015, Mazzariol et al., 2012). Furthermore, it was suggested that PCB transfer from mother to calf may cause a reduction in first-born survival rates in common bottlenose dolphins (*Tursiops truncatus*) from the northern Adriatic Sea (Genov et al., 2019).

The striped dolphin (*Stenella coeruleoalba*) is the most common cetacean species in the Mediterranean Sea (Panigada et al., 2017). Despite its abundance, the Mediterranean subpopulation has been classified as Vulnerable by the International Union for the Conservation of Nature (IUCN) (Aguilar and Gaspari, 2012).

S. coeruleoalba has been recognized as an ocean health sentinel at the sub-basin scale (Fossi and Panti, 2017; Marsili et al., 2004). Monitoring organic pollutant concentrations in this species makes it possible to oversee temporal and geographical trends, and thus, to implement future mitigation measures and renewed actions to reduce environmental contamination in the Mediterranean Sea, which have been defined as a global PCB hotspot for marine mammals (Jepson et al., 2016; Marsili et al., 2018). Moreover, the frequency of dolphin morbillivirus occurrence in this subpopulation (Mira et al., 2019; Pautasso et al., 2019) strenghtens the significance of continuous monitoring of striped dolphin strandings to figure out the role of environmental contaminants on their health status.

With the goal to contribute to the investigation on the ecotoxicological status of Mediterranean striped dolphins, the aim of this study was to assess the concentrations and congener profiles of dioxin-like polychlorinated biphenyls (dl-PCBs), PBDEs, and PCDD/Fs in blubber of striped dolphins stranded along the Italian coast. This study updates and complements the limited existing data on striped dolphins in this area, which have historically been focused on PCB marker congeners.

2. Materials and methods

2.1. Sampling

Blubber samples from 10 stranded Mediterranean striped dolphins (5 males and 5 females) were collected between 2015 and 2016 along the coasts of Italy (Fig. 1).

Detailed information on sampling and biometric data of animals can be found in Table 1. According to the carcass classification proposed by Geraci and collegues (2005), the individuals were assigned to the Code 2 (fresh animals) or Code 3 (decomposed, but organs basically intact) categories. All samples were taken from the dorsal region, anterior to the dorsal fin (Becker et al., 1999), wrapped in aluminium foil and stored at −20 °C until analysis.

2.2. Analysis of POPs

2.2.1. Sample treatment

Samples were analyzed following the procedure described in Bartalini et al. (2019). Briefly, quantities of approximately 1.5 g of blubber were lyophilized in an Edwards freeze drier for 24 h. Each sample was spiked with ($^{13}C_{12}$) surrogates of PCDD/Fs, dl-PCBs and PBDEs prior to Soxhlet extraction for 24 h using pre-cleaned cellulose thimbles and a mixture of n-hexane:dichloromethane (9:1, v:v). The lipid content of each sample was determined gravimetrically. Extracts were purified using the automated sample preparation system DEXTech + (LCTech GmbH, Dorfen, Germany). Final extracts were evaporated, transferred to vials and reduced to almost dryness under a nitrogen stream. Samples were reconstituted in nonane solutions of ($^{13}C_{12}$) surrogates of PCDD/Fs, dl-PCBs and PBDEs as internal standards for instrumental analysis. Full analytical procedure details can be found in the Supplementary data.

2.2.2. Instrumental determination

17 PCDD/Fs, 12 dl-PCBs and 27 PBDEs were identified and quantified by gas chromatography-high-resolution mass spectrometry (GC-HRMS) using a Trace GC Ultra gas chromatograph (Thermo Fisher Scientific, Milan, Italy) coupled to a high-resolution mass spectrometer (DFS, Thermo Fisher Scientific, Bremen, Germany). Different experimental conditions were established for the analysis of each family of contaminants. A full description of the instrumental parameters can be found in the Supplementary data.

2.3. QA/QC

Quality control and quality assurance measures included the analysis of blank samples covering the entire analytical procedure. All metal and glassware material was carefully precleaned (3×) using three solvents of decreasing polarity (acetone, dichloromethane and n-hexane). Quantification was based on the isotopic dilution technique in agreement with the following criteria: (a) ratio between the two monitored ions within ± 15% of the theoretical value, and (b) limits of quantification (LOQs) corresponding to a signal-to-noise ratio (S/N) of 10. Obtained values were recovery and blank corrected. The trueness of the analytical method was confirmed by means of the satisfactory analysis of the certified standard reference material SRM 1945 ("Organics in Whale Blubber", NIST). Comprehensive information related to QA/QC including surrogate recoveries, limits of detection (LODs) for target compounds, and repeatability results are provided in the Supplementary data.

2.4. Data analysis

Concentrations are expressed in ng/g (dl-PCBs and PBDEs) or pg/g (PCDD/Fs) on a lipid weight (l.w.) basis. 2,3,7,8-TCDD equivalents (TEQs) were calculated for PCDD/Fs and dl-PCBs using the World Health Organization (WHO)-2005 toxic equivalency factors (TEF) (Van den Berg et al., 2006). Concentrations below detection limits were considered as LOD values (upper bound TEQ) for PCDD/Fs and dl-PCBs. Graphics and Figures were generated using R version 3.6.2 (R Core Team, 2019) and RStudio version 1.1.463 (RStudio Team, 2016), and QGIS 3.0 Girona (QGIS Development Team, 2018).

3. Results and discussion

3.1. Levels of POPs studied

The three study POP families (dl-PCBs, PBDEs, PCDD/Fs) were detected in all striped dolphin blubber samples analyzed. All specimens showed the same pattern of relative abundance for the target contaminants: dl-PCBs > PBDEs ≫ PCDD/Fs. Six PBDE congeners (# 3, 7,

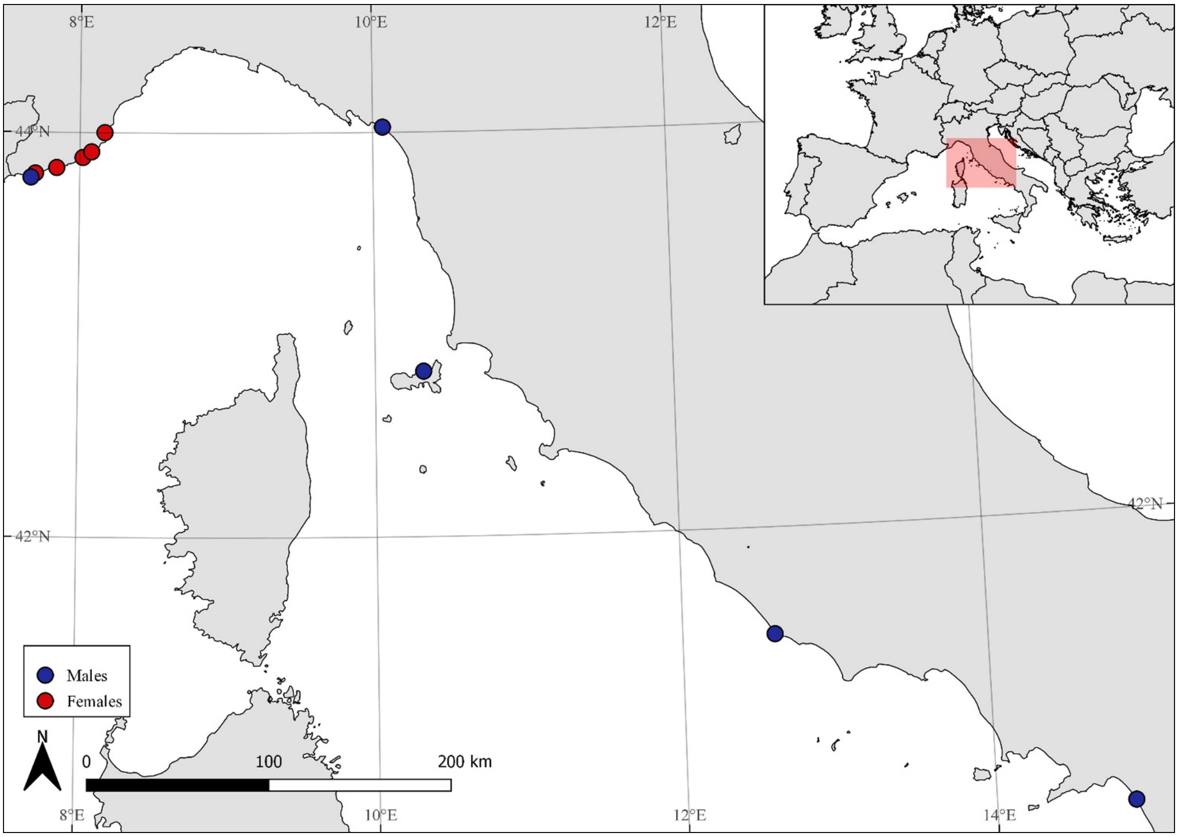

Fig. 1. Map of the sampling area.

15, 17, 119, 126) were consistently below LODs in all samples. Concentrations together with detection frequencies for each class of contaminants are shown in Table 2.

The limited sample size and the unbalance existing in the age of individuals between males and females prevented statistical analysis of sex differences. Median dl-PCB concentration was 1820 ng/g l.w. (Table 2) with males showing higher levels than females (Fig. 2A). Adult females exhibited the lowest dl-PCB concentrations (474 and 948 ng/g l.w. for Sc3 and Sc4, respectively) among all samples, while the highest levels (3840 ng/g l.w.) were recorded in a male calf (Sc7). Cetaceans' reproductive discharge, resulting in lower levels of lipophilic pollutants in females as compared to males of the same age, has already been well described (Aguilar et al., 1999; Genov et al., 2019; Reijnders et al., 2018; Reijnders et al., 2009). In striped dolphin, mothers can transfer 4%–9% and 72%–91% of their POP load to offspring during pregnacy and lactation, respectively (Murphy et al., 2018). Despite data regarding non dioxin-like PCB congeners on striped dolphin are quite abundant in literature, data on dl-PCBs –the most toxic ones– are instead fairly scarce (Law, 2014). The few data available are shown in Table 3. Non-*ortho* PCBs (IUPAC #77, 81, 126, and 169) levels in striped dolphins analyzed in this study (mean: 1.71 ng/g l.w.; median: 1.33 ng/g l.w.; range: 0.92–4.72 ng/g l.w.) were two order of magnitude lower than those recorded in diseased (Morbillivirus epizootics) dolphins in the 1990s (mean: 140 and 186 ng/g l.w.) (Borrell et al., 1996; Kannan et al., 1993), but higher than those obtained in free-ranging animals by Fossi and co-workers later on (mean: 0.372 ng/g

Table 1
Sampling information (year and site), type of stranding (passive: found dead; active: stranded alive) and biological data (sex, body length, weight, age, age class, body condition and lipid content) for analyzed striped dolphins (*Stenella coeruleoalba*, Sc) stranded along the coasts of Italy between 2015 and 2016. Abbreviations are as follows: M = male, F = female, C = calf, J = juvenile, A = adult.

Sample code	Sex	Year	Location	Type of stranding	Size (cm)	Weight (kg)	Age (y)[a]	Age class	Body condition[c]	% Lipid
Sc1	M	2015	Portoferraio, Livorno	Passive	155	29.5	3.9	J	2/3	65.19
Sc2	M	2015	Marina di Massa, Massa-Carrara	Passive	170	45	6.0	J	2	64.66
Sc3	F	2015	Bordighera, Imperia	Passive	205	75	15.9	A[b]	2	61.86
Sc4	F	2015	Imperia	Passive	207	76	16.8	A[b]	2	67.99
Sc5	F	2015	Imperia	Passive	153	42	3.7	J[b]	2	64.94
Sc6	F	2015	Sanremo, Imperia	Passive	86	6	0.6	C	2	60.24
Sc7	M	2016	Salerno	Passive	102	12	0.9	C	3	51.59
Sc8	M	2016	Anzio, Roma	Passive	150	35	3.4	J	2	68.80
Sc9	M	2016	Bordighera, Imperia	Passive	180	56.5	7.9	J	3	66.58
Sc10	F	2016	Alassio, Savona	Active	165	51.5	5.2	J	2	76.50

[a] Age determination through the mathematical model for striped dolphin described by Marsili et al., 2004.
[b] Maturity stage inferred from body length (Calzada et al., 1996, 1997).
[c] From Geraci et al., 2005.

Table 2

PBDE (ng/g lipid weight, l.w.), dl-PCB (ng/g l.w.), PCDD and PCDF (pg/g l.w.) concentrations, total TEQs (pg WHO-TEQ/g l.w.), and detection frequencies (% > LOQ), in blubber samples of striped dolphins from the Mediterranean Sea. Arithmetic mean, median and range are reported for all specimens together (N = 10) as well as for males (N = 5) and females (N = 5) separately, for each contaminant group analyzed.

		ng/g l.w.				pg WHO-TEQ/g l.w.			
Compounds	Sex	Mean	Median	Range	> LOQ (%)	Mean	Median	Range	% total TEQ
ΣPBDEs	Male	846	737	406–1660	100				
	Female	371	439	219–469	100				
	Total	**608**	**456**	**219–1660**					
Σnon-*ortho*-dl-PCBs	Male	2.25	1.42	0.97–4.72	100	65.5	53.8	28.1–132	42.7
	Female	1.17	1.10	0.92–1.51	100	33.1	35.5	24.0–38.8	44.9
	Total	1.71	1.33	0.92–4.72		49.3	37.5	24.0–132	43.8
Σmono-*ortho*-dl-PCBs	Male	2590	2230	1690–3840	100	77.7	66.8	50.8–115	55.0
	Female	1360	1730	473–1880	100	40.7	51.9	14.2–56.3	50.0
	Total	1970	1820	473–3840		59.2	54.6	14.2–115	52.5
Σdl-PCBs	Male	2590	2230	1700–3840	100	143	125	95.0–247	97.7
	Female	1360	1730	474–1880	100	73.9	75.9	50.4–88.3	94.9
	Total	**1980**	**1820**	**474–3840**		**109**	**91.6**	**50.4–247**	**96.3**
ΣPCDDs (pg/g)	Male	3.58	3.33	2.83–4.85	100	1.00	0.99	0.76–1.32	0.76
	Female	4.47	4.21	2.89–6.47	100	1.57	1.09	0.86–3.23	1.94
	Total	4.03	3.57	2.83–6.47		1.28	1.07	0.76–3.23	1.35
ΣPCDFs (pg/g)	Male	19.4	20.5	11.6–26.2	100	2.01	2.00	1.31–2.82	1.55
	Female	22.3	20.1	17.8–28.7	100	2.33	2.09	1.89–3.06	3.13
	Total	20.8	20.2	11.5–28.7		2.17	2.04	1.31–3.06	2.34
ΣPCDD/Fs (pg/g)	Male	23.0	23.8	14.4–31.0	100	3.01	2.99	2.07–3.98	2.31
	Female	26.8	24.1	21.3–35.2	100	3.90	3.53	2.98–6.29	5.07
	Total	**24.9**	**23.9**	**14.4–35.2**		**3.45**	**3.13**	**2.07–6.29**	**3.69**
Total TEQs [a]	Male				100	146	128	99.0–250	–
	Female				100	77.8	79.0	54.0–94.6	–
	Total					**112**	**96.8**	**54.0–250**	**–**

[a] TEF values from Van den Berg et al., 2006

wet weight w.w., Fossi et al., 2004). Greater PCB concentrations in diseased dolphins from 1990s should not surprise because the loss of nutritional status in cetaceans, eventually associated to the presence of infectious diseases, has previously been linked to a relative increase of blubber organochlorine residues load, as a result of lipid and pollutant mobilization, metabolisation and excretory processes (Aguilar et al., 1999; Jepson et al., 2005).

It is quite difficult to make comparisons with studies carried out in other geographical areas. Minh et al. (2000) analyzed only one specimen of striped dolphin in 1992 and found higher levels of dl-PCBs (1892 ng/g w.w., 3784 ng/g l.w.) than those found in this study. On the other hand, dl-PCB concentrations found here markedly exceeded current values reported in the southern Bay of Biscay (mean: 129 ng/g l.w.; range: 11.8–247 ng/g l.w., Romero-Romero et al., 2017). However, such a difference with the latter may be highly influenced by the nature of the biological matrices analyzed, since Romero-Romero et al. (2017) investigated dl-PCBs in striped dolphin muscle samples. Although hardly conclusive, these findings seemed to be in line with other studies carried out in the Mediterranean basin. Median levels of PCDD/Fs were 23.9 pg/g l.w. (Table 2); interestingly, slightly greater concentrations in

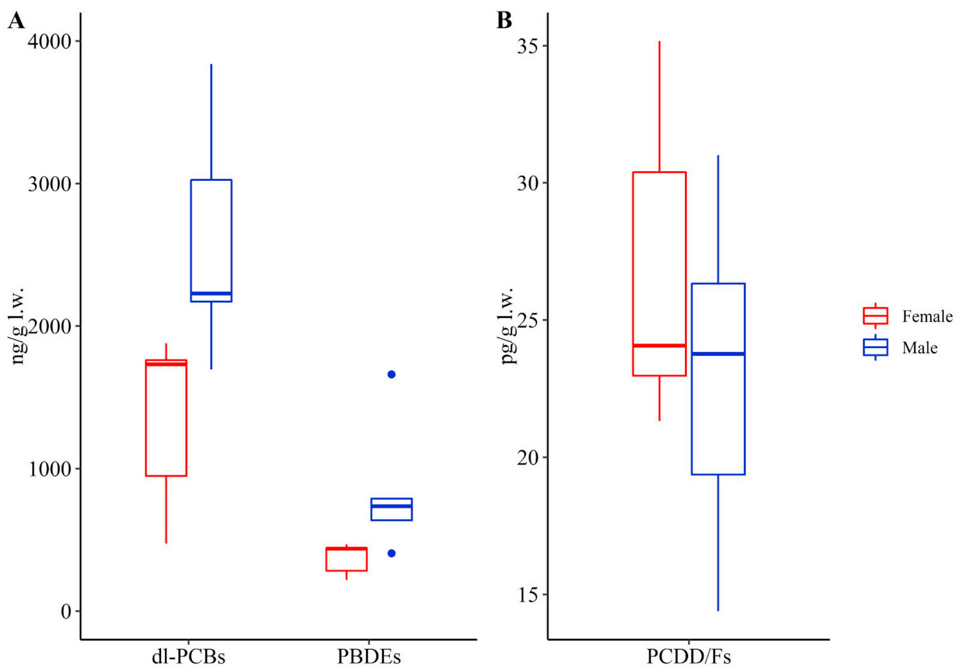

Fig. 2. Box and whisker plots of dl-PCB, PBDE (Fig. 2A, ng/g lipid weight, l.w.) and PCDD/F (Fig. 2B, pg/g l.w.) concentrations for male (N = 5, blue) and female (N = 5, red) striped dolphin (*Stenella coeruleoalba*) specimens stranded along the Italian coasts between 2015 and 2016. The dark horizontal line indicates the median and outliers are highlighted by circles. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 3

Total PCB, dl-PCB, PBDE (ng/g) and PCDD/F (pg/g) concentrations in *Stenella coeruleoalba* from different studies. Whenever possible, blubber concentrations are reported and they are expressed as mean ± SD (range) on a lipid weight basis, unless otherwise noted. Toxic equivalent quantities (TEQ) for dl-PCBs and/or PCDD/Fs are reported as pg/g l.w., unless otherwise indicated. Contaminant congeners analyzed and toxicity equivalent factors (TEF) used are noted below the Table. Whenever possible, concentrations expressed in different units in source literature were converted to ng/g or pg/g l.w. to facilitate comparisons. Summary statistics were obtained from text or tables of cited sources, or calculated from raw data reported in tables. Geographic area and year of sampling, together with number of samples, sex and lipid content (mean ± SD) are also reported when possible. Abbreviations are as follows: M = male, F = female, N = newborn, Y = young, N.D. = sex Not Determined.

Reference	Geographic Area	Sampling year	N	Sex	lipid %	ΣPCBs	Σdl-PCBs	ΣPBDEs	ΣPCDD/Fs	ΣTEQs
Mediterranean Sea										
(Cocumelli et al., 2018) [d]	Italy (Tyrrhenian Sea)	2015–2016	2 [a]	M	75.7 ± 10.3	148,523 ± 25,120 (130,760–166,285)				
(Jepson et al., 2016) [e]	Western Mediterranean Sea	1990–2009	220 [a, b]	M	–	482,150 (16,800–2,668,640)				
				F	–	312,290 (5090–1,518,640)				
(Barón et al., 2015b) [f]	Alboran Sea	2004–2011	11 [a]	8 M/3 F	–			940 (100–2250)		
(Fossi et al., 2013)[g]	Italy	2007	20 [b]		–	17,996 ± 7268		101 ± 89.2 ■		
			13 [b]		–	36,671 ± 9196		129 ± 94.8 ▲		
			14 [b]		–	72,810 ± 16,417		173 ± 113 ●		
(Storelli et al., 2012) [h]	Italy (Adriatic Sea)	1999–2004	17 [a]	9 M	43.3	39,374 ± 16,989 ∴				120,205 ‡
				6 F		5269 ± 3781 ∴				19,942 ‡
				2 N.D.		11,700 ± 5042 ∴				20,991 ‡
(Wafo et al., 2012) [i]	France	2007–2009	37 [a]	19 M	–	57,724 ± 41,900				
				13 F	–	45,315 ± 45,689				
				5 Y	–	77,198 ± 57,395				
(Castrillon et al., 2010) [j]	Spain	2004–2009	28 [a]		71.0 ± 14.3	57,170 ± 36,604 (7330–152,540)				
(Wafo et al., 2005) [i]	France	1989–1990	5 [a, c]	2 M/3 F	–	45,200 (2700–83,200)				
	France	2000–2003	3 [a, c]	1 M/2 F	87.2	(43,838–110,343)				
(Aguilar and Borrell, 2005) [k]	Spain	1987–2002	186 [b]		–	199,000 ± 150,000				
(Borrell and Aguilar, 2005) [l]	Spain (Alboran Sea)	1992–1994	20 [c]		71 ± 11	54,000 ± 27,000 (11,000–107,000)				
		1992	7 [b]		53 ± 3	109,000 ± 41,000 (62,000–167,000)				
(Pettersson et al., 2004) [m]	Italy (Tyrrhenian and Ligurian Sea)	1990	5 [a,†]	3 M	9.3 ± 1.5			5532 ± 4167 (726–8133)		
				2 F	5.0 ± 4.2			764 ± 897 (129–1398)		
(Fossi et al., 2004) [n]	Italy (Aeolian Sea)	2002	8 [b]		–		0.372 ∴		73.9 ‡	18 ‡
(Jiménez et al., 2000) [o]	Italy (Tyrrhenian and Ligurian Sea)	1990	5 [a,†]	3 M	17				75.8 ± 34.1 (39.9–108) ‡	426 ± 359 (31.6–733) ‡
				2 F	12				83.8 ± 40.5 (55.2–112) ‡	34.8 ± 11.3 (26.8–42.8) ‡
(Troisi et al., 2001) [p]	Western Mediterranean Sea	1990–1992	12 [a]		40.8 ± 4.9	75,110 ± 14,060 (34,600–142,800)				
(Reich et al., 1999) [q]	Italy (Tyrrhenian and Ligurian Sea)	1989–1990	6 [a,†]	4 M/2 F	15.1	8746 ± 11,749 ∴				1513 ± 1914
			3 [a]	2 M/1 F	42	35,599 ± 46,750 ∴				3990 ± 5986
(Marsili and Focardi, 1997) [r]	Italy	1987–1994	64 [a]	33 M/ 26 F/ 5 N.D.	74 ± 20	151,878 (6903–1,345,910) *				
(Marsili and Focardi, 1996) [r]	Italy (Ligurian, Tyrrhenian and Ionian Seas)	1991–1993	89 [b]		88.1 ± 16.1	15,500–86,000 *				
(Borrell et al., 1996) [s]	Spain	1990–1992	30 [a]	13 M/17 F	–	855,900 ± 569,000	186			
(Aguilar and Borrell, 1994) [t]	Spain	1990	72 [a]		–	778,000 (median)				
		1987–1991	109 [b]		–	282,000 (median)				

(*continued on next page*)

Table 3 (*continued*)

Reference	Geographic Area	Sampling year	N	Sex	lipid %	ΣPCBs	Σdl-PCBs	ΣPBDEs	ΣPCDD/Fs	ΣTEQs
(Kannan et al., 1993) [u]	Spain	1990	10 [a]	9 M/ 1 F	41 ± 19	1,200,000 (210,000–2,600,000)	9610–48,820			18,430 (5433–30,276) [‡]
Atlantic Ocean										
(Romero-Romero et al., 2017) [v]	Cantabrian Sea	2012–2013	2 [a,‡]		8.3 ± 8.1	2174 ± 2992 (58.3–4290)	129 ± 166 (11.8–247)	58 ± 78 (2.6–113)	2.5 ± 1.9 (1.2–3.8)	
(Méndez-Fernandez et al., 2014) [w]	Northwestern Iberian Peninsula	2008–2014	15 [a, c]	8 M	61.1 ± 19.1	22,800 ± 23,300				
				7 F		7600 ± 5900				
(Dorneles et al., 2010) [x]	Rio de Janeiro, Brazil	1994–2006	1 [a,†]	N.D.	–			210		
(Johnson-Restrepo et al., 2005) [y]	West Coast of Florida	1994	1 [a]	F	18.6	51,700		660		
(Law et al., 2003) [z]	England and Wales	1992–1998	1 [a, c]		39			450 [∴]		
Pacific Ocean										
(Bachman et al., 2014) [α]	Hawaiian Islands	1997–2010	6 [a]	3 M	37 ± 10	13,160 ± 7098 (8260–21,300)		197 ± 91 (95–269)		
				3 F	30 ± 13	14,513 ± 8942 (6240–24,000)		318 ± 169 (128–452)		
(Isobe et al., 2009) [β]	Gogo-shima Island, Ehime, Japan	2003	6 [a]	5 M	52.6 ± 5.7	32,800 ± 6420 (26,000–42,000)		632 ± 133 (520–850)		
				1 F	56.2	3200		84		
	Taiji, Japan	1978–1992	15 [c]	M	57.2 ± 15.6	22,800 ± 5680 (11,000–31,000)		270 ± 255 (13–660)		
(Marsh et al., 2005) [χ]	Japan	1999	1 [c]		–			50.8		
(Minh et al., 2000) [δ]	Sanriku, Japan	1992	1	M	50		1892			280 [‡]

[a] Stranded animals.
[b] Free ranging animals.
[c] By-caught or caught animals.
[d] Sum of PCB congeners n. 95, 99, 101, 118,128, 135, 138, 141, 144, 146, 149, 151, 153, 156, 170, 171, 172, 174, 177, 178, 180, 183, 187, 194, 195, 196, 199, 201, 202, 206.
[e] Sum of PCB congeners n. 95, 101, 110, 118, 128, 136, 138, 141, 144, 149, 151, 153, 170, 171, 174, 177, 180, 183, 187,195, 201, 202, 203.
[f] Sum of PBDE congeners n. 28, 47, 99, 100, 153, 154.
[g] Sum of PCB congeners n. 95, 99, 101, 118, 128, 135, 138, 141, 144, 146, 149, 151, 153, 156, 170, 171, 172, 174, 177, 178, 180, 183, 187, 194, 195, 196, 199, 201, 202, 206; Sum of PBDE congeners n. 17, 28, 47, 66, 85, 99, 100, 153,154, 183, 184, 191, 196, 197, 209.
[h] Sum of PCB congeners n. 8, 20, 28, 35, 52, 60, 77, 101, 105, 118, 126, 138, 153, 156, 169,180, 209; TEQs contribution from dl-PCB congeners n. 77, 105, 118, 126, 156, 169 (TEF from Van den Berg et al., 2006).
[i] The total amount of PCBs (tPCBs) was estimated as tPCBs = (CB118 + CB138 + CB153 + CB180) x 100/41.
[j] Sum of PCB congeners n. 95, 101, 103, 110, 118, 128, 136, 138, 141, 144, 149, 151, 156, 159, 170, 171, 174, 177, 180, 183, 187, 195, 201, 202.
[k] Sum of PCB congeners n. 95, 101, 110, 128, 135, 136, 138, 141, 144, 149, 151, 153, 170, 171, 174, 177, 180, 183, 187, 194, 195, 196, 201, 202, 203.
[l] Sum of PCB congeners n. 95, 101, 110, 135, 136, 138, 144, 149, 151, 153, 170, 171, 174, 177, 180, 183, 187, 194, 196, 201, 202.
[m] Sum of PBDE congeners n. 47, 99, 100, 138,153, 154, 1 unidentified PeBDE, 1 unidentified HxBDE.
[n] Sum of dl-PCB congeners n. 77, 126, 169; Sum of PCDD/F congeners 2378-TCDD, 12378-PeCDD, 123,478-HxCDD, 123,678-HxCDD, 123,789-HxCDD, 1,234,678-HpCDD, OCDD, 2378-TCDF, 12378-PeCDF, 23,478-PeCDF, 123,478-HxCDF, 123,678-HxCDF, 234,678-HxCDF, 123,789-HxCDF, 1,234,678-HpCDF, 1,234,789-HpCDF, OCDF; Sum of WHO-dl-PCB and PCDD/F–TEQ (TEF from Van den Berg et al., 1998).
[o] Sum of PCDD/Fs congeners 2378-TCDD, 12378-PeCDD, 123,478-HxCDD, 123,678-HxCDD, 123,789-HxCDD, 1,234,678-HpCDD, OCDD, 2378-TCDF, 12378-PeCDF, 23,478-PeCDF, 123,478-HxCDF, 123,678-HxCDF, 234,678-HxCDF, 123,789-HxCDF, 1,234,678-HpCDF, 1,234,789-HpCDF, OCDF; Sum of WHO-PCDD/Fs–TEQ and WHO-dl-PCBs–TEQ from Reich et al., 1999 (TEF from Van den Berg et al., 1998).
[p] Sum of 20 PCB congeners.
[q] Sum of PCB congeners n. 77, 84, 88, 91, 95, 101, 105, 110, 114, 118, 123, 126, 129, 131, 132, 135, 136, 138, 139, 144, 149, 153, 156, 157, 167, 169, 170, 171, 174, 175, 176, 178, 180, 183, 194, 196, 197; TEQs contribution from PCB congeners n. 77, 126, 169, 105, 118, 156, 157, 170, 180 (TEF from Ahlborg et al., 1994).
[r] Sum of PCB congeners n. 95, 99, 101, 118, 128, 135, 138, 141, 144, 146, 149, 151, 153, 156, 170, 171, 172, 174, 177, 178, 180, 183, 187, 194, 195, 196, 199, 201, 202, 206.
[s] Sum of dl-PCB congeners n. 77, 126, 169 and PCB congeners n. 95, 99, 101, 105, 110, 118, 128, 135, 138, 141, 144, 146, 149, 151, 153, 156, 158, 170, 171, 172, 174, 177, 178, 180, 183, 187, 193, 196, 201, 202, 203.
[t] Sum of PCB congeners n. 4, 6, 7, 8, 9, 15, 16, 17, 18, 19, 20, 24, 28, 31, 32, 33, 34, 37, 40, 41, 42, 44, 47, 49, 51, 52, 53, 58, 60, 64, 66, 67, 68, 69, 70, 72, 74, 82, 83, 84, 85, 87, 90, 91, 92, 95, 97, 99, 101, 102, 105, 110, 113, 117, 118, 120, 128, 129, 130, 132, 133, 134, 135, 136, 137, 138,,141, 144, 147, 149, 15 l, 153, 156, 159, 170, 172, 173, 176, 177, 178, 180, 183, 185, 187, 194, 195, 196, 198, 200, 201, 202, 206, 207, 208.
[u] Sum of dl-PCBs IUPAC# 77, 105, 118, 126, 156, 169 and PCB congeners n. 4, 6, 7, 8, 9, 15, 16, 17, 18, 19, 20, 24, 28, 31, 32, 33, 34, 37, 40, 41, 42, 44, 47, 49, 51, 52, 53, 58, 60, 64, 66, 67, 68, 69, 70, 72, 74, 82, 83,

84, 85, 87, 90, 91, 92, 95, 97, 99, 101, 102, 110, 113, 117, 120, 128, 129, 130, 132, 133, 134, 135, 136, 137, 138,,141, 144, 147, 149, 15 l, 153, 159, 170, 172, 173, 176, 177, 178, 180, 183, 185, 187, 194, 195, 196, 198, 200, 201, 202, 206, 207, 208; TEQs contribution from PCB congeners n. 77, 126, 169, 60, 105, 118, 156, 137, 138, 153, 170, 180, 194 (TEF from Safe, 1990).

[v] Sum of dl-PCB congeners n. 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, 189 and non dl-PCB congeners n. 28, 52, 101, 138, 153, 170, 180, 194; Sum of PBDE congeners n. 17, 28, 47, 66, 85, 99, 100, 153, 154, 184, 183, 191, 196, 197, 209; Sum of PCDD/F congeners 2378-TCDD, 12378-PeCDD, 123,478-HxCDD, 123,678-HxCDD, 123,789-HxCDD, 1,234,678-HpCDD, OCDD, 2378-TCDF, 12378-PeCDF, 23,478-PeCDF, 123,478-HxCDF, 123,678-HxCDF, 234,678-HxCDF, 123,789-HxCDF, 1,234,678-HpCDF, 1,234,789-HpCDF, OCDF.

[w] Sum of PCB congeners n. 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.

[x] Sum of PBDE congeners n. 28, 47, 66, 85, 99, 100, 153, 154, 183.

[y] Kanechlor (KC300, 400, 500, and 600) was used for the identification of PCB congeners; external calibration standards (di- through deca-CB congeners) were used for quantification of PCB congeners. Sum of PBDE congeners n. 28, 47, 66, 85, 99, 100, 138, 153, 154, 183, 203, 209, and 1 unidentified TeBDE.

[z] Sum of PBDE congeners n. 28, 47, 66, 71, 75, 77, 85, 99, 100, 119, 138, 153, 190.

[α] Sum of PCB congeners n. 8, 18, 28, 29, 31, 44, 45, 49, 50, 52, 56, 63, 66, 70, 74, 79, 82, 87, 92, 95, 99, 101, 104, 105, 106, 107, 110, 112, 114, 118, 119, 121, 127, 128, 130, 132, 137, 138, 146, 149, 151, 153, 154, 156, 157, 158, 159, 163, 165, 166, 167, 170, 172, 174, 175, 176, 177, 178, 180, 183, 185, 187, 188, 189, 191, 193, 194, 195, 196, 197, 199, 200, 201, 202, 203, 205, 206, 207, 208, 209; Sum of PBDE congeners n. 17, 25, 28, 30, 33, 47, 49, 66, 71, 75, 85, 99, 100, 116, 119, 138, 153, 154, 155, 156, 181, 183, 190, 191, 203, 205, 206, 209.

[β] Analyzed PCB congeners not reported. Sum of PBDE congeners n. 3, 15, 28, 47, 99, 100, 153, 154, 183, 196, 197, 206, 207, 209.

[χ] Sum of PBDE congeners n. 47, 99, 100, 153, 154.

[δ] Sum of dl-PCB congeners n. 77, 105, 118, 126, 156, 169; TEQs contribution from dl-PCB congeners n. 77, 105, 118, 126, 156, 169 (TEF from Van den Berg et al., 1998).

[†] Liver samples.

[‡] Muscle samples.

■ Low levels of contaminants.

▲ Moderate levels of contaminants.

● High levels of contaminants.

[**] ng/g w.w.

[‡‡] pg/g w.w.

[*] ng/g dry weight.

females than in males were measured on average (Fig. 2B). Higher values of PCDD/Fs were reported for Mediterranean striped dolphins in previous studies from 2000 (mean: 79.0 pg/g w.w., Jimenez et al., 2000) and 2004 (mean: 73.9 pg/g w.w., Fossi et al., 2004) (Table 3). More recently, Romero-Romero et al. (2017) assessed the total PCDD/F burden in muscle samples of striped dolphins from the northeastern Atlantic Ocean, finding however substantially lower levels (mean: 2.5 pg/g l.w.; range: 1.2–3.8 pg/g l.w.).

Regarding PBDEs, the median value was 456 ng/g l.w., and similarly to dl-PCBs, the highest PBDE concentrations were observed in males (Fig. 2A), and in the Sc7 specimen (1660 ng/g l.w.) in particular. The two adult females showed the lowest PBDE concentrations (219 and 283 ng/g l.w. for Sc3 and Sc4, respectively) among all samples. These values were slighty lower, but still comparable with results obtained by Barón and co-workers in the Alboran Sea (mean: 940 ng/g l.w.; range: 100–2250 ng/g l.w., Barón et al., 2015b). Compared to those previously reported for Mediterranean striped dolphins, the PBDE concentrations in the present study were higher than those recorded in free-ranging striped dolphins in the early 2000s (mean: 130 ng/g l.w., Fossi et al., 2013), but lower than levels obtained in liver samples from stranded specimens in 1990–1992 by Pettersson and colleagues (mean: 3625 ng/g l.w.; range: 129–8133 ng/g l.w., Pettersson et al., 2004) (Table 3). Worldwide, mean PBDE levels reported for striped dolphins were generally lower than those found in this study. When comparing only males, mean concentrations (Σ_{14}BDE congeners evaluated in Isobe et al., 2009) found here (mean: 820 ng/g l.w.; range: 390–1602 ng/g l.w.) approximately doubled the levels detected in specimens collected in Japan between 1978 and 2003 (mean: 362 ng/g l.w.; range: 13–850 ng/g l.w., Isobe et al., 2009) (Table 2). Likewise, the Σ_5BDE predominant congeners in biota (#47, 99, 100, 153, 154) (Law et al., 2006; Wenning et al., 2011) here was roughly one order of magnitude higher than values obtained by Marsh et al. (2005) in one Japanese specimen. Furthermore, concentrations reported for Hawaiian Islands dead striped dolphins fell well below the levels reported in the present investigation for Mediterranean striped dolphins (Bachman et al., 2014). As with the other contaminants analyzed, muscle samples from Cantabrian Sea dolphins showed a minor degree of contamination from PBDEs (Romero-Romero et al., 2017). On the other side, data gathered from the analysis of a single adult female dead along the west coast of Florida in 1994 and one specimen stranded in UK around the same time period, revealed similar or higher PBDE total amounts in the blubber compared to those found in this study (Johnson-Restrepo et al., 2005; Law et al., 2003). The greater blubber concentrations of PBDEs found in this study relative to those from other geographical areas suggest that the Mediterranean basin may be considered a PBDE hotspot, as already stated for organochlorine compunds (Jepson et al., 2016). This hypothesis is further supported by the higher PBDE body burden in striped dophin' prey from the Mediterranean Sea as compared to those from the western and eastern Pacific Ocean. The species is considered an opportunistic feeder (Aguilar, 2000), generally exploiting a wide variety of prey, typically small-sized (up to 200–300 mm length), pelagic, schooling and vertically migrating organisms. Common consumed species are cephalopods and fishes, and crustaceans to a lesser extent (Dede et al., 2016; Meissner et al., 2012; Miyazaki et al., 1973; Perrin et al., 2008; Ringelstein et al., 2006; Voliani et al., 2012; Wurtz and Marrale, 1993). PBDE levels have been investigated in deep-sea fishes from the Blanes Canyon, off the NW Mediterranean coast of the Iberian Peninsula, by Koenig et al. (2013). Ranges reported for species belonging to families *Alepocephalidae*, *Macrouridae* and *Moridae* were between 15 and 500 ng/g l.w. (0.20–3.02 ng/g w.w.). In a comprehensive study by Takahashi et al. (2010) along the Pacific coast of northern Japan, samples of *Macrouridae*, *Moridae*, *Gadidae*, *Zoarcidae*, *Myctophidae* families showed PBDE concentrations of 1.3–8.5 ng/g l.w. (0.14–0.26 ng/g w.w.). Even lower concentrations (2.5 and 5.8 ng/g l.w., i.e. 0.01–0.05 and 0.05–0.59 ng/ w.w., for *Myctophum nitidulum* and *Tarletonbeania crenularis* respectively) have been reported for myctophids from the Eastern Pacific Ocean (Gassel and Rochman, 2019). For comparison purposes, estimated lipid contents were used to convert l.w. in w.w. concentrations in Takahashi et al. (2010), by applying the equation (Eq. 10–7) provided by the U.S. Environmental Protection Agency (EPA) (2011) in the Exposure Factors Handbook.

In comparison to other Mediterranean odontocetes with similar trophic levels but distinct prey consumption and habitats, differences emerge depending on each POP family. Thus, dl-PCBs concentrations in sperm whale blubber from the Adriatic Sea (Italy) ranged from 2100 to 20,800 ng/g l.w. (Bartalini et al., 2019) being up to 5-fold greater than the highest amount found in this study (3840 ng/g lw), which underpins the still high abundance of PCBs found in the Mediterranean. On the other hand, striped dolphin PBDE levels appeared to be similar to those in common dolphins, sperm whales and Risso's dolphins (Barón et al., 2015a; Bartalini et al., 2019; Pinzone et al., 2015). PCDD/F concentrations in this study were also quite comparable to those found in sperm whales by Bartalini et al. (2019), although one order of magnitude lower that those reported in the same species by Pinzone et al. (2015). Furthermore, it could be worth citing results gathered by Barone and colleagues after the analysis of twenty-six muscle samples of Mediterranean bluefin tuna (*Thunnus thynnus*) for comparison purposes, since its trophic position is comparable with that of the striped dolphin (Sarà and Sarà, 2007; Varela et al., 2018). Tuna PCDD/F concentration levels ranged from 5.3 to 70.1 pg/g l.w. (Barone et al., 2018), being in the same order of magnitude than those obtained in this study.

3.2. Pollutant profiles

Overall, the dl-PCB congener profile was dominated by mono-*ortho* congeners, which on average accounted for 99.9% of the total burden (Figs. 3, S2).

The most prominent contibution was given by penta- and hexachlorinated congeners in all samples analyzed. Specifically PCB118 was the most abundant compound (236–1970 ng/g l.w.), representing 49.8–63.6% of total dl-PCBs, followed by PCB105 (13.4–19.5%), PCB156 (6.2–14.7%) and PCB167 (8.8–11.9%) (Fig. 3). These findings are in accordance with dl-PCB profiles previously reported in striped dolphins, and in other marine mammals (Dorneles et al., 2013; Gaus et al., 2005; Helm et al., 2002; Moon et al., 2010; Storelli et al., 2012; Vijayasarathy et al., 2019).

Conversely, altogether the non-*ortho* PCB congeners gave a negligible contribution (0.1% on average) to the total concentration of dl-PCBs (Figs. 3, S2). The mean residue levels were found to be in the order PCB77 (0.38–2.36 ng/g l.w.) > PCB169 (0.24–1.19 ng/g l.w.) > PCB126 (0.13–0.96 ng/g l.w.). This profile did not follow commercial mixture patterns (PCB77 > PCB126 > PCB169), yet the same results were documented for highly contaminated Mediterranean odontocetes in the 1990s (Corsolini et al., 1995; Kannan et al., 1993). Among the three coplanar congeners, PCB169 is the most stable, while PCB126 and PCB77 are relatively more biodegradable (Kannan et al., 1993). High PCB tissue loads have been associated with an enhanced induction of mixed function oxidase (MFO) activity in striped dolphins (Fossi et al., 1992; Tanabe and Tatsukawa, 1992); thereby, resulting in the metabolism of more biodegradable congeners. Kannan and coworkers, found a linear relationship between PCB169:PCB126 ratio and total PCB concentration in striped dolphins affected by the morbillivirus epizootic (Kannan et al., 1993). Even though no correlation between Σdl-PCBs and coplanar congener ratios was found in samples of this study -possibly due to the low number of individuals analyzed- a heightened detoxifying activity may account for the relative higher levels of hexachlorinated to that of pentachlorinated congeners found in this study. In order to explain the predominance of the most biodegradable PCB77 among three non-*ortho* coplanar congeners a higher exposure than elimination rate can be suggested owing to its greater presence in commercial mixtures (Agency for Toxic Substances and Disease Registry (ATSDR), 2000; Kannan et al., 1987). PCB77 was found dominant as well by Fossi et al. (2004) and Minh et al. (2000) in Mediterranean striped dolphins.

TetraBDEs were the most abundant congeners among all PBDEs, with BDE47 ranging from 39 to 560 ng/g l.w., in line with BDE47 concentrations found in striped dolphins from the Alboran Sea (Barón et al., 2015b) (Figs. 4, S3).

Important contributions were also given by BDE154 (6 Br), BDE100 (5 Br), BDE99 (5 Br), which accounted for 22%, 14% and 11% on average of total PBDEs, respectively (Fig. S3). Despite the widespread use in Europe of the DecaBDE commercial mixture (Hites, 2004; La Guardia et al., 2006; Lee and Kim, 2015), trace levels of BDE209 were detected only in 3 out of 10 striped dolphins of this study. This result was somewhat expected given the low bioavailability of this congener in marine environments owing to its high affinity to particle binding (Lee and Kim, 2015), its debromination under abiotic conditions (Salvadó et al., 2012), and its short-life due to biotransformation in biota (Ross et al., 2009; Zhang et al., 2016). Contribution of BDE154 to total PBDEs in all specimens analyzed was higher than data previously reported for Mediterranean striped dolphins, and more generally for marine mammals worldwide (Bachman et al., 2014; Pettersson et al., 2004; Zhang et al., 2016). Nevertheless, this result is in agreement with findings by Isobe et al., (2009) in adult striped dolphins from Japan. Upon closer examination, not all striped dolphins shared an identical PBDE congener profile (Fig. 4), contrary to what was found for dl-PCBs. All males displayed the same congener pattern for the most abundant PBDEs: BDE47 > BDE154 > BDE100 > BDE99. Conversely, this pattern seemed to vary with age in females. Younger female specimens showed a clear prevalence of low-brominated congeners -TetraBDE and PentaBDE- with BDE47 alone reaching almost 60% of total PBDEs in the calf Sc6. Tetra-BDE relative contribution gradually declined in older specimens (BDE47 accounted for 41% and 39% in Sc5 and Sc10, respectively), as long as it represented only a small fraction of the total in older females (BDE47 accounted for 18% and 23% in Sc3 and Sc4, respectively). In adult females, HexaBDE (namely, BDE154 and BDE153) were the most abundant congeners, contributing up to 55% to the total PBDE load. Given the low number of samples, this relative enrichment of highly brominated congeners associated with achievement of sexual maturity could be explained simply by dissimilar metabolic efficiencies and elimination capacities between individuals. Alternatively, it could be assumed that low-brominated, thus less lipophilic and low-molecular weight, congeners may be more easily discharged during pregnancy and lactation (Desforges et al., 2012; Kajiwara et al., 2008). A similar hypothesis was put forward by Kajiwara et al. (2008) and Desforges and co-workers (2012) for melon-headed whales (*Peponocephala electra*) and arctic beluga whales (*Delphinapterus leucas*), who found significant inverse correlations between log Kow and mother-fetus ratios of PBDE congeners.

Concerning PCDD/Fs, contribution of PCDFs (84%) was roughly 4-fold greater than that of PCDDs (16%) in total (Figs. 5, S4).

This was quite different from what was described by Fossi et al. (2004) and Jiménez et al. (2000) in Mediterranean striped dolphins who observed the same contribution from PCDDs and PCDFs. Overall, the most abundant congeners were 1,2,3,4,7,8-HxCDF (0.85–10.6 pg/g l.w.), 2,3,7,8-TCDF (1.69–4.95 pg/g l.w.) and 2,3,4,7,8-PeCDF (1.73–3.67 pg/g l.w.), accounting for 19%, 14% and 10% of total PCDD/Fs on average, respectively (Fig. S4). All dioxin congeners had a contribution under 5%; among them the most abundant compunds were 1,2,3,7,8-PeCDD (0.48–2.62 pg/g l.w., 3.7% of PCDD/Fs), followed by OCDD (0.41–1.74 pg/g l.w., 3.2% of PCDD/Fs) and 1,2,3,4,6,7,8-HpCDD (0.36–1.26 pg/g l.w., 2.9% of PCDD/Fs). Despite slight inter-individual variability, both PCDD and PCDF fingerprints were dominated by hexa- and penta-substituted congeners (Fig. 5). Contrary to previous findings (Fossi et al., 2004; Jiménez et al., 2000), higher chlorinated congeners gave a negligible contribution to total, while lower chlorinated congeners were major contributors of PCDD/F

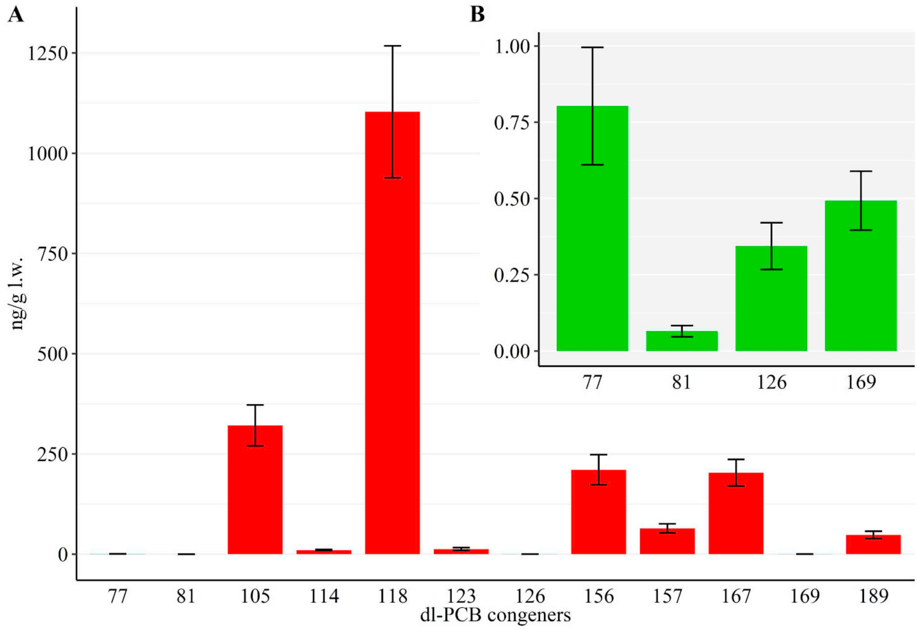

Fig. 3. Average contribution of individual mono-*ortho* (red) and non-*ortho* (green) PCB congeners to the total dioxin-like PCB burden (ng/g l.w.) in Mediterranean striped dolphins ($N = 10$). Error bars represent standard error (SE). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

profiles. A reduced bioaccumulation and biomagnification of highly chlorinated PCDD/Fs in marine ecosystems has been cogitated and widely described in literature (Khairy et al., 2014; Ruus et al., 2006; Strid et al., 2007; Wan et al., 2005). For instance, higher concentrations of tetra- and penta-PCDD/Fs compared to hexa-to octa-PCDD/Fs has been reported for Mediterranean top predators like fish-eating birds, turtles, sharks and bluefin tunas (Roscales et al., 2016; Sprague et al., 2012; Storelli et al., 2011; Storelli and Zizzo, 2014). In this study, the OCDD abundance represented the most striking difference related to previous investigations referring to this congener as the greatest contributor to PCDD/F profiles (Fossi et al., 2004; Jiménez et al., 2000). In some of those studies the occurrence of higher chlorinated compounds was associated with local combustion sources (Jiménez et al., 2000). Therefore, it could be suggested that the abundance pattern found in this study may respond to a change, or a reduction, in dioxins and furans environmental sources over a time period of two-three decades. Similarly, a decreasing trend in the relative proportion of OCDD was found for ringed seals from East Greenland between 1999 and 2003 (Riget et al., 2005).

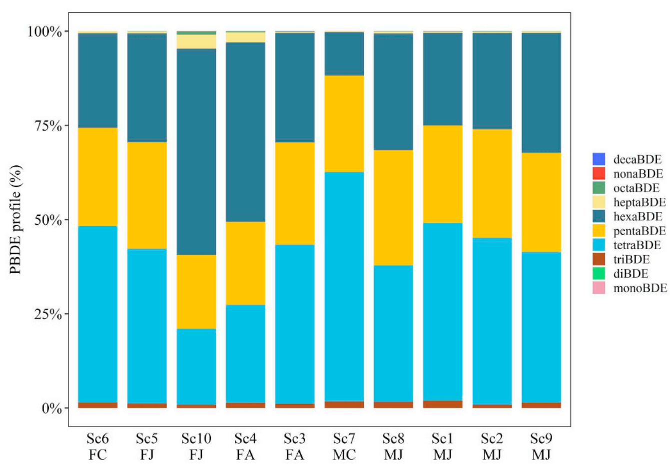

Fig. 4. PBDE congeners profile (%) in striped dolphin (*Stenella coeruleoalba*) specimens ($N = 10$) stranded along the Italian coasts between 2015 and 2016.

3.3. Toxicity assessment

Polychlorinated dibenzo-*p*-dioxins, dibenzofurans, and dioxin-like PCBs are structurally related aryl hydrocarbon receptor (AhR)-active compounds, exhibiting the same specific toxic mode of action (Sorg, 2014). Their toxic potential can be measured in fractional equivalencies of TCDD (2,3,7,8-tetrachlorodibenzo-*p*-dioxin), the most toxic and best studied member of its class, in order to facilitate risk assessment and regulatory control (Sanderson and Van den Berg, 1999; She et al., 2016). Using the toxic equivalent factors (TEFs) for mammals developed by Van den Berg et al. (2006), TEQs were calculated for all study dolphins. Total TEQ values ranged from 54.0 to 250 pg/g l.w. (Fig. 6).

Most of the specimens exhibited TEQs below the threshold value of 210 pg WHO-TEQ/g l.w., associated with immunological dysfunctions in harbor seals (Ross et al., 1995), while only one juvenile male (specimen Sc7) exceeded that value (Fig. S5). It should be underlined, however, that the use of these estimates entails some degree of uncertainty due to species-specific susceptibility to environmental pollutants. The great variability in compounds analyzed, TEFs used (Ahlborg et al., 1994; Safe, 1990; Van den Berg et al., 2006, 1998), together with diverse analytical methods and types of samples, make it difficult to compare data over time (Table 3). The most recent study addressing TEQs in Mediterranean striped dolphins reported values that were remarkably higher than those found in this study (Storelli et al., 2012). Prior to this, other authors reported TEQ levels that were consistent with what found here (Fossi et al., 2004; Jiménez et al., 2000). TEQs reported in this study were below values found in Mediterranean sperm whales, pilot whales and bottlenose dolphins (Bartalini et al., 2019; Pinzone et al., 2015; Romanić et al., 2014; Storelli et al., 2007; Storelli and Marcotrigiano, 2003).

The average TEQ profile was noticeably dominated by dl-PCBs, which accounted for 93–99% of total TEQs (Fig. 7).

Despite concentrations of mono-*ortho* PCBs substantially exceeded those of non-*ortho* congeners in all specimens, their TEQ contribution was fairly similar on average. Therefore, the TEQ pattern was somewhat inter-individually variable, as shown in Fig. S5. PCDD/Fs provided only a small contribution to total TEQs (1.2–6.7%). Except for specimen Sc5, in all cases the contribution of PCDFs to total TEQs was higher than that of PCDDs. Major contributions were given by PCB126 (30.5%), followed by PCB118 (29.4%) and PCB169 (13.2%). Among PCDD/Fs, 1,2,3,7,8-PeCDD and 2,3,4,7,8-PeCDF provided the highest

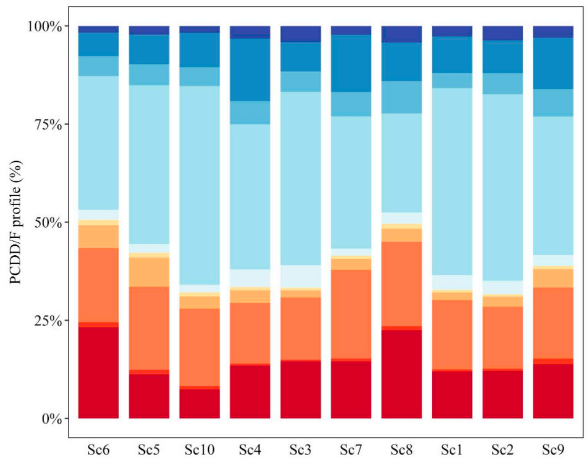

Fig. 5. PCDD/F congeners profile (%) in striped dolphin (*Stenella coeruleoalba*) specimens ($N = 10$) stranded along the Italian coasts between 2015 and 2016.

Fig. 7. PCDD/F and dl-PCB relative contribution profile (%) to total TEQs in striped dolphin (*Stenella coeruleoalba*) specimens (N = 10) stranded along the Italian coasts between 2015 and 2016.

contribution to TEQs. The predominance of dl-PCBs was in agreement with what had been already reported for striped dolphins (Marsili et al., 2018), and more generally for other cetaceans in the Mediterranean Sea and worldwide (Dorneles et al., 2013; Pinzone et al., 2015).

Potential health risks associated with the exposure to PBDEs in mammals include thyroid disruption, neurodevelopmental and reproductive toxicity (Alonso et al., 2014; Kodavanti et al., 2018). Almost all dolphins studied here showed PBDE values below the upper limit of the threshold level (1500 ng/g l.w.) associated with thyroid endocrine disruption in juvenile grey seals (Hall et al., 2003). Noteworthy, as for TEQs, the PBDE load found in blubber of the specimen Sc7 (1660 ng/g l.w.) surpassed this threshold (Fig. S6).

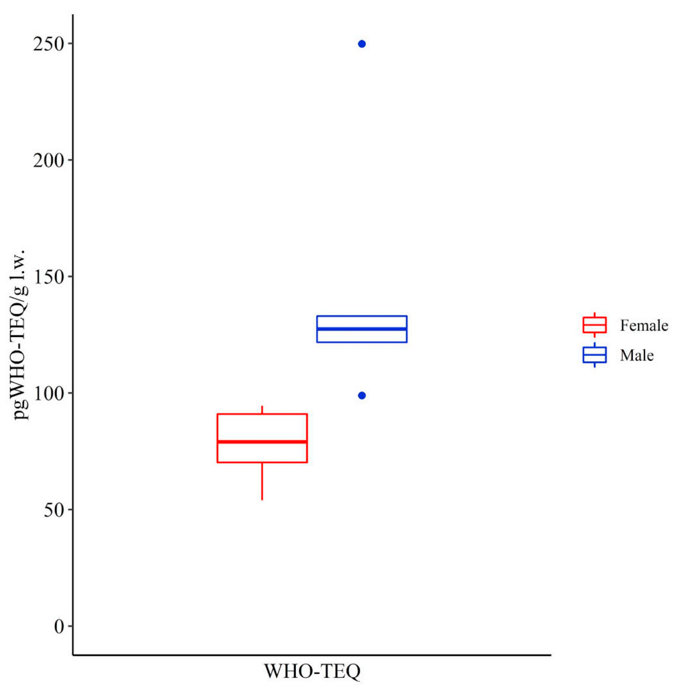

Fig. 6. Box and whisker plots of dl-PCB, PCDD and PCDF total WHO-TEQ concentrations (pgWHO-TEQ/g l.w.) for male ($N = 5$, blue) and female (N = 5, red) striped dolphins (*Stenella coeruleoalba*) specimens stranded along the Italian coasts between 2015 and 2016. The dark horizontal line indicates the median and outliers are highlighted by circles. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

4. Conclusions

Despite the restrictions imposed globally, the present study emphasized that POP exposure levels in the striped dolphins from the Mediterranean Sea have not declined significantly in recent years. In particular, the higher PBDE concentrations found here compared to other geographical areas may advocate for a new role of the Mediterranean Sea as a pollution hotspot for organobromine compounds, beyond the same already recognized role for oganochlorine contaminants. In spite of the EU ban on their production and use in the mid-1980s, PCBs still make the major contribution to the total toxic equivalency (TEQ), thereby representing the main source for possible health impairment. This scenario highlights the persistent need to monitor such pollutants in the Mediterranean Sea where high levels of POP exposure in marine mammals have been documented in order to provide scientific evidence for their associated toxicological risk, which in turn, may help to implement new strategies of mitigation and reduction of environmental contamination.

CRediT authorship contribution statement

Francesca Capanni: Formal analysis, Investigation, Data curation, Writing - original draft, Visualization.**Juan Muñoz-Arnanz:**Methodology, Validation, Data curation, Writing - review & editing, Supervision.**Letizia Marsili:**Conceptualization, Resources, Writing - review & editing, Supervision, Project administration.**M. Cristina Fossi:**Conceptualization, Writing - review & editing, Supervision.**Begoña Jiménez:**Conceptualization, Methodology, Resources, Writing - review & editing, Supervision, Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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