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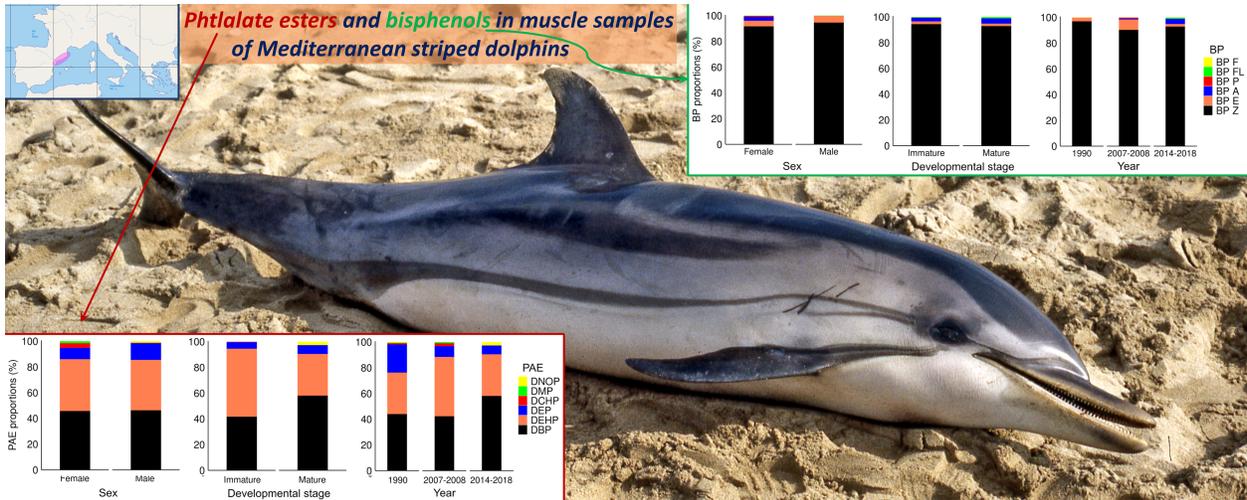
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**CRedit author statement**

**Morgana Vighi:** Conceptualization, Investigation, Writing - Original Draft, Visualization, Supervision; **Asunción Borrell:** Conceptualization, Methodology, Resources, Writing - Review & Editing, Supervision; **Wissam Sahyoun:** Methodology, Formal analysis; **Sopheak Net:** Methodology, Resources, Writing - Review & Editing, Supervision; **Alex Aguilar:** Conceptualization, Resources, Writing - Review & Editing, Supervision; **Baghdad Ouddane:** Methodology, Resources; **Odei Garcia-Garin:** Conceptualization, Methodology, Formal analysis, Investigation, Data curation, Writing - Review & Editing, Visualization, Project administration, Funding acquisition.

Journal Pre-proof



1 **Concentrations of bisphenols and phthalate esters in the muscle of Mediterranean striped**  
2 **dolphins (*Stenella coeruleoalba*)**

3

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12

13 **Highlights**

14 .

- 15 1. Muscle samples were obtained from 30 stranded Mediterranean striped dolphins.  
16 2. Six bisphenols and 6 phthalate esters were detected in the samples.  
17 3. BPZ was detected in all the samples, at the highest concentrations.  
18 4. Concentrations did not relate with biological traits but varied with time.  
19 5. Results confirm dolphin exposure to plasticizers in an already anthropized basin.

20

21 **Abstract**

22 Bisphenols (BPs) and phthalate esters (PAEs) are important compounds for the plastics industry, also  
23 called “everywhere chemicals” due to their ubiquity in daily use products. Both chemical groups are  
24 well-known environmental contaminants, whose presence has been reported in all environmental

25 compartments, and whose effects, mainly associated to endocrine disruption, are detrimental to living  
26 organisms. Cetaceans, due to their long life-span, low reproduction rate and high position in the trophic  
27 web, are especially vulnerable to the effects of contaminants. However, little is known about BP and  
28 PAE concentrations in cetacean tissues, their potential relation to individual biological variables, or  
29 their trends over time. Here, the concentration of 10 BPs and 13 PAEs was assessed in the muscle of  
30 30 striped dolphins (*Stenella coeruleoalba*) stranded along the Spanish Catalan coast (NW  
31 Mediterranean) between 1990 and 2018. Six BP and 6 PAE compounds were detected, of which only  
32 4,4'-(cyclohexane-1,1-diyl)diphenol (BPZ) was detected in all the samples, at the highest  
33 concentration (mean 16.06  $\mu\text{g g}^{-1}$  lipid weight). Sex or reproductive condition were largely uninfluential  
34 on concentrations: only dimethylphthalate (DMP) concentrations were significantly higher in  
35 immature individuals than in adults, and the overall PAE concentrations were significantly higher in  
36 males than in females. Temporal variations were only detected in bis(4-hydroxyphenyl)ethane (BPE),  
37 diethylphthalate (DEP) and dimethylphthalate (DMP), whose concentrations were lower, and 9,9-  
38 Bis(4-hydroxyphenyl)fluorene (BPFL), which were higher, respectively, in samples taken between  
39 2014-2018, probably reflecting shifts in the production and use of these chemicals. These results  
40 provide the first assessment of concentrations of several BP and PAE compounds in the muscle of an  
41 odontocete cetacean.

42

### 43 **Keywords**

44 Pollutants; Plastic additives; Plasticizers; Contamination; Marine Mammals; Cetaceans

## 45 1. Introduction

46 Bisphenols (BPs) and phthalic acid esters (PAEs) have been called “everywhere chemicals”  
47 due to their widespread use in the manufacture and processing of plastic and resin products and their  
48 ubiquity in human body, wildlife and the environment (Net et al, 2015; Warner and Flaws, 2018;  
49 Sangeetha et al, 2021). Indeed, they are important chemical components for the plastics industry, where  
50 BPs (and more specifically Bisphenol A (2,2-bis(4-hydroxyphenyl)propane, henceforth BPA), along  
51 with its analogues) are used in the manufacture of hard, transparent plastics such as water bottles, as  
52 well as in the production of polycarbonate, polyacrylate, flame retardants, solvents, and other products  
53 used for coating, such as epoxy resins, and are present in a large variety of everyday products (Staples  
54 et al, 1998; Rochester et al, 2015; Mustieles et al, 2020; Torres-García et al, 2022). On the other hand,  
55 PAEs are used to manufacture soft, flexible and transparent plastics, such as plastic polyvinyl chloride  
56 (PVC) products, and a very broad range of products including building materials, toys, personal care  
57 products (PCP), food packaging, cosmetics, medical devices (Graham, 1973), and many other daily  
58 use products, where their content can be up to 10-60 % by weight (Net et al, 2015).

59 Although plastic materials are chemically inert, the chemical additives used to improve their  
60 qualities may be released to the environment during the plastic weathering processes caused by solar  
61 radiation, sea corrosion, and chemical and biological degradation (Koelmans et al, 2014; Net et al,  
62 2015). Moreover, BPs and PAEs may also enter the environment during the production process and  
63 ultimately be transported at sea through river discharge or atmospheric deposition (Xie et al., 2007).  
64 The constant raise of plastic production, and consequent plastic pollution in the worldwide oceans  
65 (Jambeck et al, 2015; Geyer et al, 2017; Lambert et al, 2020) is thus expected to increase the  
66 concentration of plastic additives such as BPs and PAEs in the marine environment, as a result of their  
67 direct deposition, and/or their leaching from the large amounts of marine plastic litter that reach the  
68 oceans every day.

69 Indeed, both BPs and PAEs are well-known environmental pollutants, which have been  
70 detected in every natural compartment, including air, soil, sediments, fresh and marine waters (Xie et  
71 al, 2007; Zeng et al., 2009; Ben Sghaier et al, 2017; Paluselli et al, 2018; Moustafa et al, 2022; Torres-  
72 García et al, 2022; Xing et al, 2022), as well as in the tissues of several animal species (Kang et al,  
73 2007; Koelmans, 2014; Nehring et al, 2017; Routti et al., 2021). Both *in vivo* and *in vitro* studies have  
74 proven their toxic effects to living organisms. *In vitro* studies with laboratory animals and  
75 epidemiological studies in humans showed that BPA, several of its analogues, and most PAEs, cause  
76 severe endocrine effects (Basak et al, 2020), as well as potential neurological and carcinogenic effects  
77 (e.g., Benjamin et al, 2017; Li et al, 2018; Pelch et al, 2019; Han et al, 2021). Although most BPs and  
78 PAEs are subject to efficient metabolic degradation and elimination (Kang et al, 2006; Net et al, 2015),  
79 the increased levels of environmental and dietary exposure to which living organisms are subject may  
80 maintain their tissue concentrations at chronic or sub-chronic levels, and trigger detrimental effects  
81 (Silva et al, 2018; Warner and Flaws, 2018).

82 Cetaceans are particularly vulnerable to the effects of lipophilic contaminants. Being mammals  
83 with a long life-span, low reproduction rate, and high position in the trophic web, they are subject to  
84 both the bioaccumulation and biomagnification of pollutants (Loganathan, 2012). Despite the proven  
85 toxic effects of BPs and PAEs to mammals, and their potentially high concentrations in the marine  
86 environment, their concentrations in the tissues of marine mammals have been determined only by a  
87 handful of studies so far. Most of the published research focuses on the assessment of PAEs and their  
88 metabolites in the tissues of two main species: the bottlenose dolphin (*Tursiops truncatus*, *i.e.*, Dziobak  
89 et al, 2021, 2022; Hart et al, 2018, 2020) and the fin whale (*Balaenoptera physalus*, *i.e.*, Bains et al,  
90 2017; Fossi et al, 2012, 2014, 2016; Garcia-Garin et al, 2022); while other marine mammal species  
91 have been scarcely studied (e.g., Rian et al, 2020; Routti et al, 2021), and the concentrations of BPs in  
92 these organisms have been barely investigated (*i.e.*, Montoro-Martínez et al, 2021; Page-Karjian et al,  
93 2020). Still little is known regarding the concentrations of these contaminants, and especially of BPs

94 and PAEs other than the more commonly studied BPA and di-(2-ethylhexyl) phthalate (DEHP), in  
95 marine mammals, their potential relation with biological variables, or their trends over time.

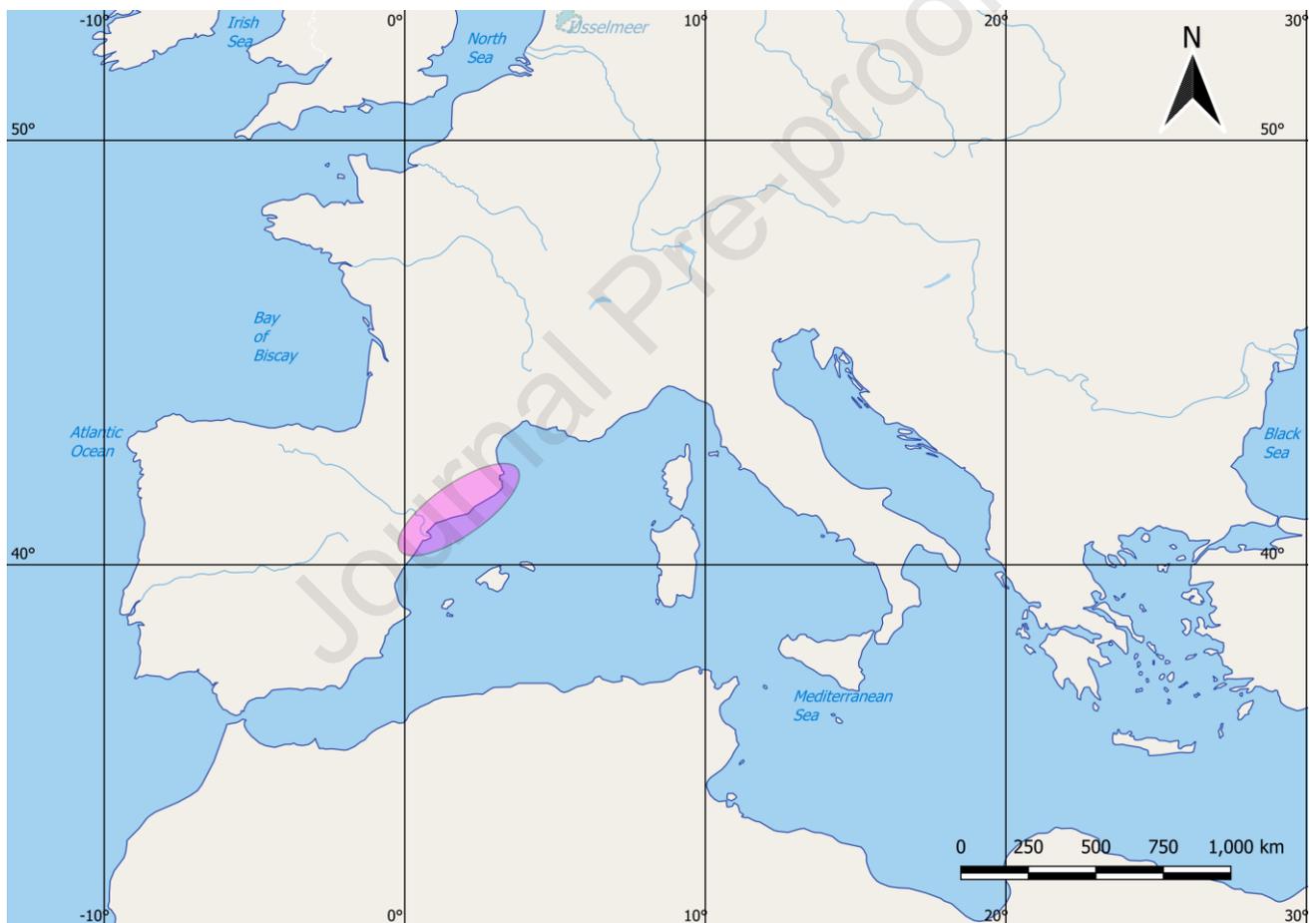
96 With the aim of contributing to fill such information gap, here we analysed 10 BPs and 13 PAEs  
97 in the muscle of 30 striped dolphins (*Stenella coeruleoalba*) stranded along the Mediterranean coast  
98 of Spain between 1990 and 2018. Our results are discussed taking into account the potential influence  
99 of biological variables, such as sex and maturation stage of the individuals, and of the year of death,  
100 which might reflect the increasing levels of plastic pollution in the Mediterranean Sea and the  
101 regulation of some of the analysed compounds, like BPA and DEHP. Although the Mediterranean  
102 striped dolphin is currently not in a concerning conservation status, cetaceans are often subject to a  
103 variety of human impacts that, combined, may compromise the healthy survival of their populations  
104 (Hart et al, 2020, Dziobaak et al, 2022). The pathways and levels of exposure to these increasingly  
105 widespread pollutants of potentially vulnerable marine fauna such as cetaceans deserve in-depth  
106 investigation.

## 107 **2. Materials and methods**

### 108 *2.1. Study area and sampling*

109 Muscle samples were collected from 30 striped dolphins found stranded along the Spanish  
110 Catalan coast, in the NW Mediterranean Sea (Fig.1), during three separate periods between 1990 and  
111 2018 (*i.e.*, 1990, 2007-08, and 2014-18), and preserved in the biological tissue bank of the University  
112 of Barcelona (BMA Tissue Bank). Since muscle represents approximately 50% of dolphins weight  
113 and its average lipid content is 6%, this tissue was considered representative enough to assess the levels  
114 of BPs and PAEs in the dolphins. The cause of death for most dolphins was related with the  
115 morbillivirus epizootics occurred in the Mediterranean Sea during 1990 and 2006-07 periods. As age  
116 information was only available from 7 out of the 30 dolphins, individuals were classified according to

117 their sexual maturation stage, defined according to their body length. Individuals whose body was  
118 shorter than 1.3 m were classified as immature, and individuals whose body was longer than 1.8 m  
119 were classified as adult (Table 1). Overall, sampled individuals included 24 adults (12 males and 12  
120 females), and 6 immature individuals (2 males and 4 females, Table 1). Muscle samples, stored at  $-20$   
121  $^{\circ}\text{C}$ , were subsampled using stainless steel material to obtain individual subsamples of 25-30 g, which  
122 were placed in glass bottles and preserved frozen until analysis. To avoid contamination, no plastic  
123 material was used during this procedure.



124  
125 Figure 1: Map of the Western Mediterranean Sea highlighting (in pink) the area of the Spanish Catalan coast, where the  
126 striped dolphins sampled in this study were found stranded.

127

128 Table 1: Sex, body length (in m), maturation stage and age (when determined) of the sampled striped dolphins, along with  
 129 the lipid content and overall phthalate and bisphenol compounds concentrations found in their muscle (expressed in  $\mu\text{g g}^{-1}$   
 130 lipid weight (lw)).

Sample	Year	Sex	Length	Maturation stage	Age	Lipid content (%)	$\Sigma$ PAE	$\Sigma$ BP
2007_1	2007	M	2.1	MATURE	NA	1.24	72.499	1.277
2007_3	2007	F	1.8	MATURE	NA	1.21	9.564	17.251
2007_4	2007	F	1.95	MATURE	NA	0.69	30.430	32.939
2007_6	2007	M	1.85	MATURE	NA	1.10	23.231	17.249
2008_1	2008	F	2.1	MATURE	NA	15.46	3.568	0.369
2008_6	2008	F	1.89	MATURE	NA	1.08	46.250	26.104
2008_7	2008	M	1.85	MATURE	NA	1.04	7.923	42.594
9033	1990	M	1.93	MATURE	16	1.87	26.645	2.714
9034	1990	F	1.96	MATURE	14	1.64	3.542	8.971
9040	1990	M	1.81	MATURE	7	1.77	14.713	13.499
9042	1990	M	1.87	MATURE	10	1.67	4.463	50.728
9043	1990	F	1.8	MATURE	9	1.85	1.547	4.665
9046	1990	F	1.82	MATURE	30	1.58	5.192	9.069
9054	1990	F	2	MATURE	23	1.77	5.020	17.188
9067	1990	M	1.91	MATURE	NA	1.86	36.006	28.954
N_1_18	2018	F	2	MATURE	NA	1.04	2.089	1.905
N_168_17	2017	M	1.93	MATURE	NA	1.30	18.476	5.756
N_21_18	2018	M	1.82	MATURE	NA	1.40	15.992	0.924
N_286_15	2015	F	1	IMMATURE	NA	2.88	13.259	10.305

N_291_14	2014	F	1.04	IMMATURE	NA	16.78	1.832	0.154
N_352_14	2014	M	1.3	IMMATURE	NA	1.39	0.006	15.661
N_355_14	2014	F	1.07	IMMATURE	NA	3.10	9.681	33.913
N_370_14	2014	M	2.04	MATURE	NA	1.42	4.522	2.240
N_407_15	2015	M	2.1	MATURE	NA	1.22	6.141	20.854
N_45_17	2017	F	1.23	IMMATURE	NA	3.02	80.258	13.801
N_488_17	2017	M	1.98	MATURE	NA	1.38	16.826	24.986
N_49_15	2015	F	1.9	MATURE	NA	2.06	11.118	5.772
N_497_17	2017	F	1.9	MATURE	NA	8.77	1.806	3.242
N_523_17	2017	M	1.03	IMMATURE	NA	2.70	8.303	96.396
N_77_17	2017	F	1.81	MATURE	NA	1.41	1.372	4.375

131

132 *2.2. Sample preparation*

133 After 48 h lyophilization, a subsample of 4 g (dry weight, dw), was taken from the central  
134 portion (to avoid possible contamination) of each muscle sample and homogenised. Each subsample  
135 was then transferred in 50 mL centrifuge tubes and diluted in a mixture of 10 mL acetonitrile, 10 mL  
136 Milli-Q water, 100  $\mu$ L internal standard (d<sub>4</sub>-DIBP), and extraction salts (composed by 1 g NaCl, 4 g  
137 MgSO<sub>4</sub>, 1 g C<sub>6</sub>H<sub>5</sub>Na<sub>3</sub>O<sub>7</sub>·2H<sub>2</sub>O and 0.5 g C<sub>6</sub>H<sub>6</sub>Na<sub>2</sub>O<sub>7</sub>·1.5H<sub>2</sub>O). After stirring in a vortex mixer with  
138 ceramic homogenizers and centrifugating for 5 minutes at 5000 rpm, 8 mL of each extract were  
139 transferred to centrifuge tubes containing the dispersive solid phase extraction clean-up salts (400 mg  
140 PSA, 400 mg GCB, 1200 mg MgSO<sub>4</sub> and 400 mg C18EC). The mixture was then vortexed and  
141 centrifuged again at 5000 rpm for 5 minutes. Five (5) mL of the supernatant were transferred to a vial,  
142 and the extraction was repeated a second time using 10 mL ethyl acetate. The two resulting extracts  
143 were then combined and reduced under a gentle nitrogen stream to 200  $\mu$ L and 50  $\mu$ L for PAEs and

144 BPs, respectively. Prior to PAE GC-MS/MS analyses, extracts were stored frozen in glass tubes. For  
145 BPs, each extract was derivatized with 50  $\mu$ L of BSTFA containing 1% TMCS and heated at 65  $^{\circ}$ C  
146 during 2 h. After derivatization, samples were kept at room temperature during 15 min prior to GC-  
147 MS analysis (Sghaier et al, 2017).

148 Due to the lipophilic nature of PAEs and BPs, it is generally recommended to report their  
149 concentrations on a lipidic weight (lw) basis to prevent any bias related to the individual, specific, or  
150 tissue-related differences in lipid concentrations (Krahn et al, 2003). Thus, following Garcia-Garin et  
151 al (2020), 1 g dw of each sample was used to determine the lw gravimetrically by a 15 min sonication  
152 in a 15 mL solution of hexane:acetone (1:1). Two extractions were performed and the resulting extracts  
153 were mixed. After the solvent evaporation under nitrogen stream, the remaining lipids were dried at  
154 90  $^{\circ}$ C until they reached a constant weight.

### 155 2.3. Instrumental analysis, standards and reagents

156 Ten BP and thirteen PAE compounds were analysed, namely: BPA, 4,4'-(1-  
157 Phenylethylidene)bisphenol (BPAP), 1,1-Bis(4-hydroxyphenyl)-1,1-diphenylmethane (BPBP), 2,2-  
158 Bis(4-hydroxy-3-methylphenyl)propane (BPC), bis(4-hydroxyphenyl)ethane (BPE), (bis(4-  
159 hydroxyphenyl)methane (BPF), 9,9-Bis(4-hydroxyphenyl)fluorene (BPFL), 4,4'-(1,3-  
160 Phenylenediisopropylidene)bisphenol (BPM), 4,4'-(1,4-Phenylenediisopropylidene)bisphenol (BPP),  
161 4,4'-(cyclohexane-1,1-diyl)diphenol (BPZ), dimethylphthalate (DMP), diethylphthalate (DEP), di-  
162 butylphthalate (DBP), bis(2-methoxyethyl) phthalate (DMEP), dipentylphthalate (DPP), bis(2-  
163 ethoxyethyl) phthalate (DEEP), benzyl butyl phthalate (BBP), phthalic acid dicyclohexyl ester  
164 (DCHP), bis(4-methyl-2-pentyl) phthalate (BMPP), di-*n*-hexyl phthalate (DHP), bis(2-*n*-butoxyethyl)  
165 phthalate (DBEP), bis(2-ethylhexyl) phthalate (DEHP) and di-*n*-octyl phthalate (DNOP). Analytical  
166 standards of BPs and PAEs were purchased from TCI Europe (Zwijndrecht, Belgium) and Restek  
167 (Lisses, France), respectively. Deuterated BPA (BPA d16) and labelled phthalic acid diisobutyl ester

168 (d<sub>4</sub>-DIBP) were obtained from Sigma-Aldrich (St. Louis, MO, USA); acetonitrile and ethyl acetate  
169 (for HPLC isocratic grade) for trace analysis were provided by VWR; the extraction salts were  
170 provided by Merck Acetone (SupraSolv®) and Agilent Technologies; and the BSTFA + TMCS (99:1)  
171 solution was provided by Supelco.

172 An Agilent 7890 GC coupled to an Agilent 7000C Triple Quadrupole GC/MS system (Agilent  
173 Technologies, France) were used for analyses. An Agilent DB-17ms, 30 m × 0.25 mm, 0.25 μm  
174 column, with a constant flow of 1.2 mL helium (99.999% purity) min<sup>-1</sup>, was used to produce analyte  
175 separation. The solvent vent injection mode was used through an ultra-inert inlet liner, with a glass  
176 wool frit from Agilent, to inject samples in a multimode inlet. The injector operated with an injection  
177 volume of 3 μL and a temperature cycle of: 0.8 min at 50 °C (solvent evaporation), raise to 300 °C at  
178 720 °C min<sup>-1</sup>; 5 min hold; cooling down to 280 °C; 10 min hold. The quenching and collision gas were  
179 helium (99.999% purity; flow rate of 2.25 mL min<sup>-1</sup>) and nitrogen (99.999% purity; flow rate of 1.5  
180 mL min<sup>-1</sup>), respectively. The temperature cycle of the oven included: 2 min at 40 °C, a first raise to  
181 220 °C at 30 °C min<sup>-1</sup>, a second raise to 260 °C at 5 °C min<sup>-1</sup> and a last raise to 280 °C at 20 °C min<sup>-1</sup>,  
182 7 min hold; for a total run time of 24 min. Electron impact ionisation (EI) at 70 eV ionising energy,  
183 and the MRM mode were set to operate the triple quadrupole mass spectrometer, at temperatures of  
184 280 °C for the transfer line, 230 °C for the ion source and 150 °C for quadrupoles 1 and 2.

#### 185 2.4. Quality assurance

186 To prevent sample contamination, all procedures were performed in clean laboratory  
187 conditions: samples and standards preparation, extraction, clean-up and the pre-concentration step  
188 were carried out in a laminar flow cabinet. Glassware was washed with detergent (Decon), rinsed with  
189 ultra-pure water (Milli-Q water) then washed with acid solution (HCl) and again rinsed with Milli-Q  
190 water follow by acetone and was dried at 120 °C prior to use. Analytical blanks were included every  
191 10 samples to minimize the error of quantification. Targeted compounds in blanks should be lower

192 than limit of quantification (LOQ) value: if they were higher than LOQ in the samples, the  
193 concentration detected in the blank was subtracted from the corresponding batch of samples (Net et al,  
194 2015). BPs and PAEs were lower than LOQ in the procedural blanks. Limit of detection (LOD) and  
195 LOQ for each compound were estimated as 3-times and 10-times the baseline of blank chromatograms,  
196 respectively, and were confirmed by experimental analysis. LOQs for BPs and PAEs were in the range  
197 of 0.73 – 4.36 and 0.012 – 0.062 ng g<sup>-1</sup> dw, respectively. The recovery rate of each BP and PAE was  
198 calculated by spiking the targeted compounds into a real matrix of interest. The mean BP recovery was  
199 estimated at 99.7% with a rate of 108, 102, 110, 106, 94 and 97% for BPA, BPE, BPF, BPFL, BPP,  
200 and BPZ, respectively. The mean PAE recovery was estimated at 92% with a rate of 104, 81, 86, 98,  
201 76 and 106% for DMP, DEP, DBP, DEHP, DCHP and DNOP, respectively. Samples were not replicated  
202 but the analytical procedure was repeated to check its replicability: Standard deviations obtained were  
203 <5% when the concentration of the compound was higher than 0.1 ng g<sup>-1</sup> and ~12% when the  
204 concentration of the compound was close to the LOQs. Calibration curves were obtained in the range  
205 of 0.1 – 5000 µg l<sup>-1</sup> with the correlation coefficients of  $\geq 0.994$ .

206 The University of Lille, LASIRE laboratory (UMR CNRS 8516) provided the infrastructure  
207 and support to perform all above analyses.

## 208 2.5. Statistical analysis

209 The distribution of each BP and PAE concentrations were tested for normality and  
210 heteroscedasticity through a Shapiro Wilk and Levene tests, respectively. As concentrations were not  
211 normally distributed, data were analysed using non-parametric tests, considering both each BP and  
212 PAE compound individually, as well as the sum of all BP and of all PAE compounds. To exclude  
213 potential biases, results obtained from mature individuals (n=24) were used to assess the relative  
214 influence of sex and year of death, split in three groups (*i.e.*, 1990, 2007-2008 and 2014-2018), through  
215 a Kruskal Wallis and a Spearman's rho correlation test, respectively. Results obtained from the 2014-

216 2018 samples (n=15) were used to assess the potential effect of the maturation stage through a Kruskal  
 217 Wallis test. The level of significance was set at  $p < 0.05$ . Analyses were performed in R (R Core Team,  
 218 2021).

### 219 3. Results

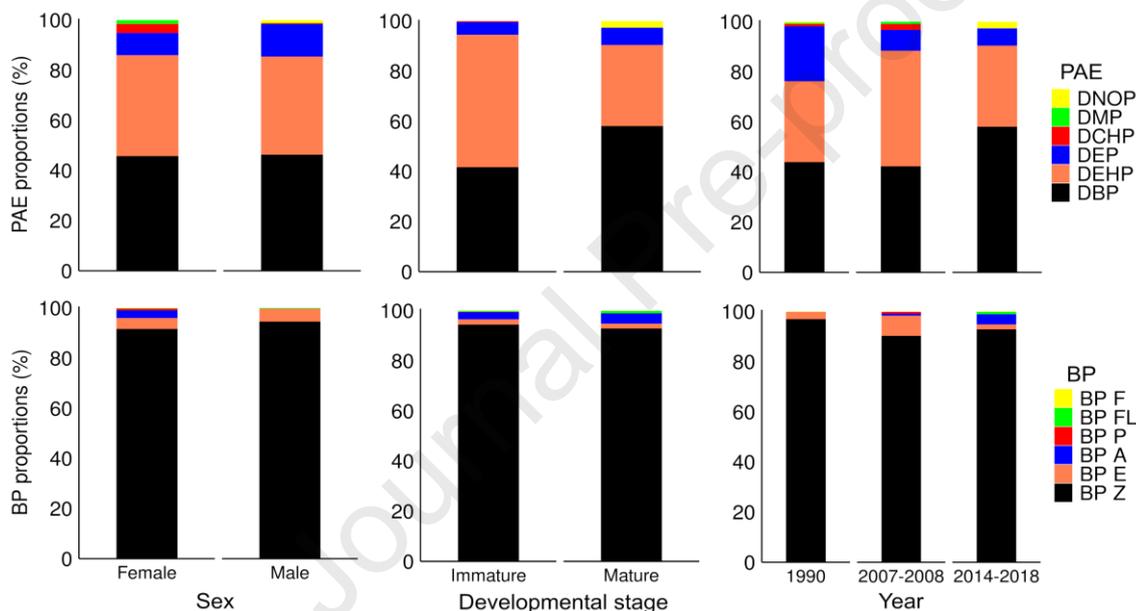
220 The overall concentration of BPs and PAEs detected in each sample are shown in Table 1.  
 221 Minimum, maximum and mean concentrations of each BP and PAE, as well as their frequency of  
 222 occurrence, are summarized in Table 2. Out of the 10 BPs and 13 PAEs analysed, 6 BPs and 6 PAEs  
 223 were detected, respectively. The concentrations of the majority of selected BPs and PAEs were below  
 224 the LOQ, meaning that these compounds were not detected in all the sampled dolphins. Indeed, BPZ  
 225 was the only compound detected in all samples, at higher concentrations than any other compound  
 226 (Table 2). DEP, DBP and DEHP were the only other compounds whose mean concentrations were  
 227 higher than  $1 \mu\text{g g}^{-1} \text{lw}$ , and which were also detected in 80%, 73% and 63% of samples, respectively  
 228 (Table 2, Fig. 2).

229 Table 2: Mean concentrations and ranges of individual and overall BP and PAE compound concentrations expressed on a  
 230 lipid weight (lw) basis, and frequency of occurrence (FO) in the muscle of stranded striped dolphins. Values below LOQ  
 231 are expressed as 0.

	Mean lw ( $\mu\text{g g}^{-1}$ )	Range ( $\mu\text{g g}^{-1} \text{lw}$ )	FO (%)
BPA	0.29	0-4.93	10
BPE	0.67	0-6.17	90
BPF	0.01	0-0.07	67
BPFL	0.05	0-0.44	47
BPP	0.03	0-0.94	7
BPZ	16.06	0.12-94.9	100
$\Sigma\text{BP}$	17.13	0.15-96.4	

DBP	7.24	0-26.59	73
DCHP	0.19	0-3.03	13
DEHP	6.85	0-48.9	63
DEP	1.62	0-8.04	80
DMP	0.08	0-1.38	38
DNOP	0.09	0-2.1	7
$\Sigma$ PAE	16.08	0.01-80.29	

232



233

234 Figure 2: Relative contribution of the detected BP and PAE compounds split by sex, maturation stage, and sampling period.

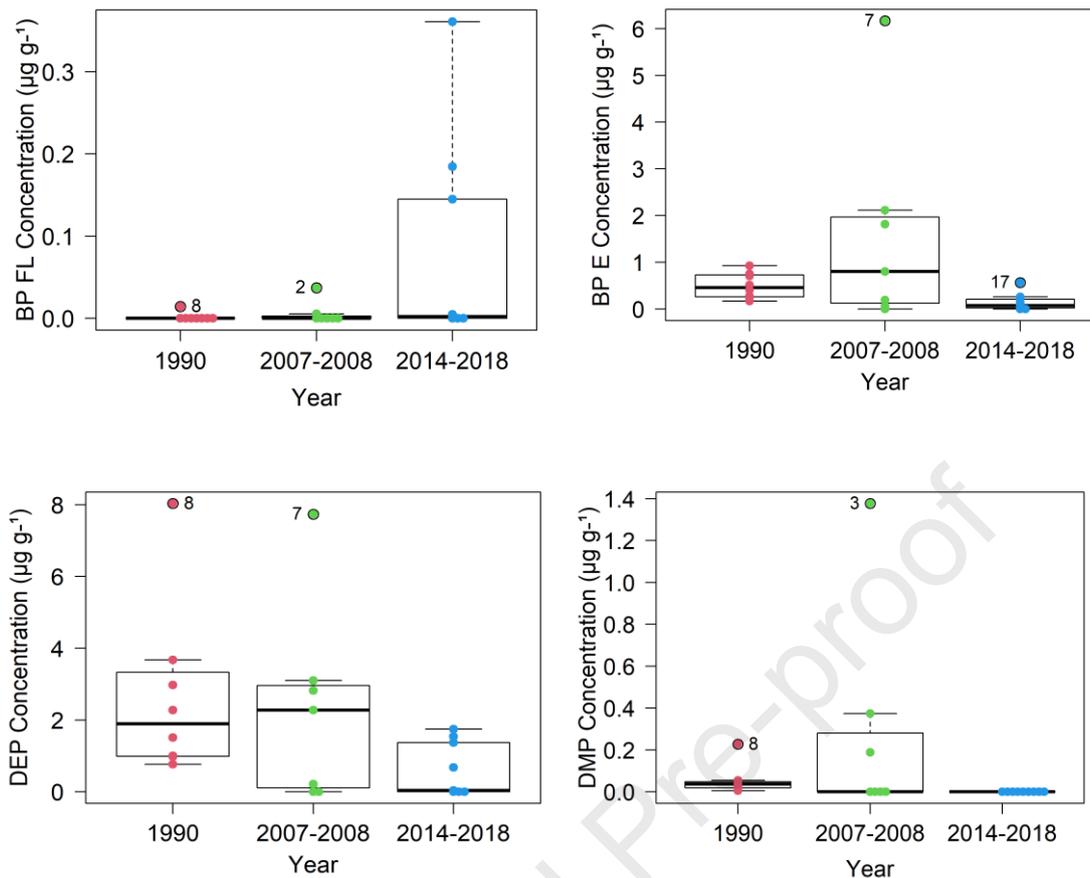
235 Results of the comparison of BP and PAE concentrations between sexes, mature and immature  
 236 individuals, and sampling periods are shown in table 3. No significant difference was detected between  
 237 sexes for individual BPs or PAEs. However, the overall concentration of PAEs was significantly higher  
 238 ( $p = 0.024$ , Kruskal-Wallis test) in samples from males than in those from females. No significant  
 239 difference was detected between the concentrations of BPs or PAEs in mature and immature dolphins,  
 240 with the exception of DMP, whose concentrations were significantly higher ( $p = 0.023$ , Kruskal-Wallis  
 241 test) in immature individuals than in adults. Finally, a significant positive correlation between the

242 concentration of BPFL and the period of sampling ( $\rho = 0.46, p = 0.024$ ), and significant negative  
 243 correlations between the concentrations of BPE, DEP, and DMP and the period of sampling were  
 244 detected ( $\rho = -0.49, p = 0.016, \rho = -0.494, p = 0.014; \rho = -0.708, p < 0.001$ , respectively) (Spearman's  
 245 rho test, Table 3, Fig. 3).

246 Table 3. Results of the statistic tests performed to compare the concentrations of BPs and PAEs between males and females,  
 247 mature and immature individuals, and the three sampling periods. The number of samples considered for each test is  
 248 indicated in brackets. K-W refers to Kruskal-Wallis test. Significant results ( $p < 0.05$ ) are marked with an asterisk (\*).

	Sex (24 samples)		Maturation Stage (15 samples)		Sampling period (24 samples)		
	K-W chi-squared	<i>p</i> -value	K-W chi-squared	<i>p</i> -value	Spearmans' S	Spearmans' rho	<i>p</i> -value
BPA	2.087	0.1486	0.15873	0.6903	1910.3	0.169422	0.4287
BPE	0.27047	0.603	3.5619	0.05912	3422.3	-0.487936	0.01557*
BPF	0.65906	0.4169	3.0247	0.08201	3022	-0.3139107	0.1352
BPFL	0.15862	0.6904	1.2763	0.2586	1245.2	0.4585894	0.0242*
BPP	1	0.3173	1.5	0.2207	2336.8	-0.01599233	0.9409
BPZ	0.65333	0.4189	2	0.1573	3056	-0.328701	0.1168
ΣBP	0.40333	0.5254	2	0.1573	3007.2	-0.3074647	0.1439
DBP	1.0967	0.295	0.057613	0.8103	2395.2	-0.04140386	0.8477
DCHP	0.4943	0.482	1.5	0.2207	2671.3	-0.1614263	0.4511
DEHP	2.3774	0.1231	0.12963	0.7188	2376.3	-0.03319363	0.8776
DEP	3.4433	0.06351	0.031818	0.8584	3435.8	-0.4938182	0.01419*
DMP	0.14256	0.7057	5.1734	0.02294*	3928.4	-0.7079887	0.0001086*
DNOP	2.087	0.1486	0.66667	0.4142	2292.3	0.003369187	0.9875
ΣPAE	5.07	0.02434*	0	1	2489	-0.08217524	0.7027

249



251

252 Figure 3: Boxplots showing the significant correlations between BPs (BPFL and BPE, top graphs) and PAEs (DEP and  
 253 DMP, bottom graphs) concentrations detected in samples from adult individuals and the period of sampling. Boxes show  
 254 the first and the third quartiles, horizontal lines represent the median, and whiskers cover extreme values within the 1.5  
 255 IQR value. Outliers are marked as dots.

#### 256 4. Discussion

257 In this study, we assess the occurrence and concentration of BPs and PAEs in striped dolphins  
 258 that stranded in the Western Mediterranean Sea across the 30-year period between 1990 and 2018, and  
 259 provide the first record, to our knowledge, of the presence of a number of these compounds in the  
 260 muscular tissue of cetaceans. We discuss our results in light of the scarce literature available on the  
 261 concentration of these contaminants in the marine environment, and specifically on marine mammals.  
 262 In the published literature, summarised in Table 4, these pollutants are assessed in cetaceans or marine  
 263 mammals through a variety of tissues, including muscle, liver, blubber, skin, plasma and urine, which

264 only allows a speculative discussion of the different values observed by other studies. Taking the  
265 limitations implied by the relatively low number of samples analysed into account, we also consider  
266 the potential influence of factors liable of influencing the detected concentrations, such as sex,  
267 maturation stage, and year of death of individuals. As information regarding the cause of death was  
268 not available for all individuals, and most of the sampled dolphins were killed by the morbillivirus  
269 epizootic, results could not be related to any potential toxic or detrimental effect to the health status of  
270 dolphins.

271 Table 4. Concentrations of BP and PAE compounds assessed in the tissues of marine mammal species. All concentrations  
272 are expressed in  $\text{ng g}^{-1}$ , with the exception of those in urine samples, which are expressed in  $\text{ng mL}^{-1}$ . The matrix in which  
273 the compounds analysed, along with the number of samples and the specification on the basis on which concentrations are  
274 calculated (dw: dry weight, ww: wet weight, lw: lipid weight) are indicated in the first column; the geographic area of each  
275 study, along with the corresponding reference are indicated in the last column.

Species	#samples, matrix	BBP	DBP	DCHP	DEHP	MEHP	DEP	MEP	DMP	DNOP	BPA	BPF	BPS	Area, reference
Fin whale ( <i>Balaenoptera physalus</i> )	5, Blubber (ww/lw) 30/10, skin + blubber biopsies (ww) 3, skin (dw) 6, Blubber (ww) 31, Muscle (dw)					58/177 55; 40								Italy <sup>a,b</sup> Mediterranean Sea/Sea of Cortez <sup>c</sup> Mediterranean Sea <sup>d</sup> Svalbard <sup>e</sup> Iceland <sup>f</sup>
Blue whale ( <i>Balaenoptera musculus</i> )	7, Blubber (ww)	<10		<10	20		<37			<10				Svalbard <sup>e</sup>
Bowhead whale ( <i>Balaena mysticetus</i> )	5, Blubber (ww)	<10		<10	n.a		<37			<10				Svalbard <sup>e</sup>
Minke whale ( <i>Balaenoptera acutorostrata</i> )	2, Liver (ww)	29.7	10.7		86.2		15.1		2.5	6.7				Greenland <sup>g</sup>
Striped dolphin ( <i>Stenella coeruleoalba</i> )	2, Skin (dw) 5, Muscle 30, Muscle (lw)				21460	1720								Mediterranean Sea <sup>d</sup> Canaries <sup>h</sup> This study
<i>Stenella spp.</i>	5, Blubber (dw)						70				<10			USA <sup>i</sup>
Risso's dolphin ( <i>Grampus griseus</i> )	1, Skin (dw) 2, Muscle				1130	464		42			163	89	37	Mediterranean Sea <sup>d</sup> Canaries <sup>h</sup>
Bottlenose dolphin ( <i>Tursiops truncatus</i> )	1, Skin (dw) 2, Muscle 46, Blubber (dw) 17/51/50, Urine (ng mL <sup>-1</sup> )		413		26068	1770		0			201	110	10	Mediterranean Sea <sup>d</sup> Canaries <sup>h</sup> U.S. <sup>1</sup> U.S., <sup>j</sup> U.S. <sup>k,1</sup> U.S. <sup>m</sup>
Pilot whale ( <i>Globicephala melas</i> )	3, Liver (ww)	28	15.1		103.6		23.2		2.50	5.6				Faroe <sup>f</sup>
Short-finned pilot whale ( <i>Globicephala macrorhynchus</i> )	1, Muscle		969		335		0				732	81	12	Canaries <sup>h</sup>
Harbour porpoise ( <i>Phocoena phocoena</i> )	100, Liver (ww)							5.99						Norway <sup>n</sup>

Pygmy sperm whale ( <i>Kogia breviceps</i> )	1, Muscle	664		102	0		181	47	213	Canaries <sup>h</sup>
<i>Kogia spp</i>	22, Blubber (dw)				170		1650			U.S. <sup>i</sup>
Fraser's dolphin ( <i>Lagenodelphis hosei</i> )	1, Muscle	552		330	98		72	67	28	Canaries <sup>h</sup>
White-beaked dolphin ( <i>Lagenorhynchus albirostris</i> )	1, Blubber (dw)				1380 0		397400			U.S. <sup>i</sup>
Melon-headed whale ( <i>Peponocephala electra</i> )	3, Blubber (dw)				500		<10			U.S. <sup>i</sup>
Polar bear ( <i>Ursus maritimus</i> )	12, Fat tissue (ww)	<10	<10	n.a.	<37.5		<10			Svalbard <sup>e</sup>
	13, Plasma (ww)				<0.8	<0.8				Svalbard <sup>e</sup>
	2, Liver (ww)	31	12.8	142.6	22.7	51.4	22			Greenland <sup>f</sup>
Ringed seal ( <i>Phoca hispida</i> )	4, Liver (ww)	128.6	7.3	117.2	21	3.7	7.6			Greenland <sup>f</sup>

276 <sup>a</sup>Fossi et al (2012);277 <sup>b</sup>Fossi et al (2014);278 <sup>c</sup>Fossi et al (2016);279 <sup>d</sup>Baini et al (2017) (also analysed MBzP, MBP, BBzP, DIOIP);280 <sup>e</sup>Routti et al (2021) (also assessed DMPP, DHxP, and DnNP, DnBP, DiBP, DiNP and DiDP in blubber of cetaceans and adipose tissue of polar bear; and

281 MMP, MEOHP, MiBP, MnBP, MBzB, MEHHP in the blood plasma of polar bear);

282 <sup>f</sup>Garcia-Garin et al (2022),283 <sup>g</sup>Vorkamp et al (2004),284 <sup>h</sup>Montoto Martinez et al (2021),285 <sup>i</sup>Page-Karjian et al (2020) (they also analysed muscle samples from *Mesoplodon spp*, *G. griseus* and *Z. cavirostris* for BPA and DEP, which were not

286 detected);

287 <sup>j</sup>Hart et al (2018) (also analysed MEOHP and MBP);288 <sup>k</sup>Hart et al, 2020 (also assessed MBP);289 <sup>l</sup>Dziobak et al (2021) (also assessed MBP, MBzP, MiBP, MEOHP, MEHHP, MMP),290 <sup>m</sup>Dziobak et al (2022);291 <sup>n</sup>Rian et al (2020) (also assessed mIBP, mBP, PA, mMP, mHxP, mEHHP, mNP, mOP, mEOHP, mHpP, mBzP, mDP).

292

## 293 4.1 BP concentrations

294 Since its commercialization in the 1950s, BPA has been the most widely used BP during decades  
295 (Staples et al, 1998). However, after several studies alerted about its adverse effects on reproduction,  
296 development, and neural networks, as well as on the cardiovascular, metabolic, and immune systems  
297 of living organisms, its use was discontinued for most applications in the European Union (EC,  
298 2011/8/EU), as well as in Canada, United States, Japan, Brazil, China and Malaysia (Akhbarizadeh et  
299 al, 2020). Industry soon after began to develop alternative compounds to replace it, and in 2016, 16  
300 BP analogues were industrially applied, of which BPF, BPS and BPAF were the most widely used  
301 (Chen et al, 2016). Due to their increased production and use, several of these compounds were soon  
302 detected in foodstuff, household dust, and PCPs, and high concentrations were detected in both the  
303 environment and human tissues (Pelch et al, 2019; Park et al, 2020).

304 In this study, we detected 6 of the 10 BPs analysed, which, in decreasing order of frequency, were:  
305 BPZ, BPE, BPF, BPFL, BPA, BPP, and, in decreasing order of concentration: BPZ, BPE, BPA, BPFL,  
306 BPP, BPF. Diet is the main intake route for marine mammals, which can ingest BPs either indirectly  
307 through contaminated prey (*i.e.*, fish or molluscs) and water, or directly from accidentally ingested  
308 pieces of plastics, microplastics, or other materials that may release BPs during their degradation  
309 (Nehring et al, 2017). In smaller proportions, marine mammals may also be exposed to these  
310 contaminants through the inhalation of polluted air (Nehring et al, 2017).

311 According to our results, BPZ was the most common and most concentrated compound (mean 16.06,  
312 range 0.12-94.9  $\mu\text{g g}^{-1}$  lw), followed by BPE (mean 0.67, range 0-6.17  $\mu\text{g g}^{-1}$  lw), which was detected  
313 in 90% of the samples, at slightly higher concentrations than the other four BP compounds detected.  
314 Unfortunately, very little is known about the environmental concentration of both compounds. BPZ is  
315 a component widely used in European countries for the manufacture of epoxy resins and polycarbonate  
316 plastics, and has been detected in PCP (0.71-0.74  $\text{ng g}^{-1}$ ), foodstuff (0.03-0.08  $\mu\text{g g}^{-1}$ ), soft drinks (0.09  
317  $\mu\text{g L}^{-1}$ ) and human biomonitoring samples (0.06  $\mu\text{g L}^{-1}$ ), as well as in indoor dust and sludge (at 0.001-

318 0.5, and up to 1.8 ng g<sup>-1</sup>, respectively, Chen et al, 2016), which may explain its higher concentration  
319 in all the analysed samples. On the other hand, BPE is used for polycarbonate and cyanate and epoxy  
320 resins, and recent studies have shown that it is commonly detected in source and drinking water (0.83  
321 ng L<sup>-1</sup>, Zheng et al, 2019), as well as in municipal sewage sludge (0.980 µg g<sup>-1</sup>, Sauer et al, 2021).  
322 Moreover, according to Danzl et al (2009), BPE in seawater is slightly more resistant to biodegradation  
323 than other compounds such as BPA, BPF or BPP, which may justify its presence in 90% of the samples  
324 analysed.

325 The other four BPs were not detected in all the samples, as their lowest concentrations were below the  
326 LOQ. Mean and maximum concentrations were below 1 µg g<sup>-1</sup> lw for all these compounds, except for  
327 BPA, which was only detected in three individuals, but whose maximum concentration was one order  
328 of magnitude higher than BPP, BPFL and two orders of magnitude higher than BPF. Indeed, although  
329 in Europe the use of BPA has been banned from several products, this compound is still widely  
330 produced and has been found in surface waters (0.25 µg L<sup>-1</sup>, Zheng et al, 2019), sewage sludge (0.12–  
331 6.50 µg g<sup>-1</sup>, Sauer et al, 2021), bivalves, holothurians and fish from the Balearic Island, in the Western  
332 Mediterranean Sea (0.29; 0.45 and 0.32 µg g<sup>-1</sup> ww, respectively, Rios-Fuster et al, 2022), samples of  
333 herring gull feathers, Baltic grey seal fur, and human hair (0.1, 0.07 and 0.34 µg g<sup>-1</sup>, respectively,  
334 Nehring et al, 2017), as well as in the few cetacean species analysed (Page-Karjian et al, 2020,  
335 Montoto-Martinez et al, 2021), which also included 5 striped dolphins (Table 4). Ozhan and Kocaman  
336 (2019) detected BPA levels in the highly anthropized waters of the Black Sea, Bosphorus, and Sea of  
337 Marmara in a range of 8.85–14.76 µg L<sup>-1</sup>. However, BPA is rapidly degraded in seawater (Staples,  
338 1998), where it can last only up to 30 days (Kang et al, 2007) and, similarly to other BPs, its excretion  
339 rate in mammals is high (Kang et al, 2006), which possibly explains its low presence in the dolphins  
340 analysed (*i.e.*, 3 out of 30).

341 BPFL was detected in almost half of the samples, at lower concentrations than all the previously  
342 mentioned compounds. This compound is a novel BPA substitute used in the plastics industry as an  
343 organic (fluorinate) synthesis intermediate (Zhang et al, 2021), and extensively applied to synthesize  
344 polyesters, epoxy resins, polycarbonates, and in consumer products that are labelled BPA-free (Özkan-  
345 Kotiloğlu et al, 2022). Aside from its detection in animal feeds (0.24 ng g<sup>-1</sup> dw, Wang et al, 2021) and  
346 in environmental samples of water bodies and sediments (0.056-0.069 ng L<sup>-1</sup>, Jin and Zhu, 2016), the  
347 information regarding this compound is very limited. However, its increasing use may be the reason  
348 of its presence in almost half of the sampled dolphins.

349 Finally, mean BPP and BPF concentrations were similarly low, but while BPP was detected only in 2  
350 samples (7%), BPF was detected in almost 70% of them. Indeed, BPF is widely applied for the  
351 manufacture of lacquers, varnishes, liners, adhesives plastics, and water pipes, as well as in dental  
352 sealants, oral prosthetic devices, tissue substitutes and coatings for food packaging (Chen et al, 2016).  
353 Both compounds have been detected in foodstuff (*e.g.*, 0.24 µg g<sup>-1</sup> BPP, and 1.13 µg g<sup>-1</sup> BPF,  
354 respectively, in samples from mustard and ginger, Liao and Kannan, 2013), household dust (up to 2 ng  
355 g<sup>-1</sup> BPP, and up to 1 µg g<sup>-1</sup> BPF, respectively, Chen et al, 2016), and PCPs (0.79-0.99 ng g<sup>-1</sup> BPP, and  
356 0.82-0.99 µg g<sup>-1</sup> BPF, respectively, Chen et al, 2016), and BPF has also been detected in sewage sludge  
357 (0-3.1 µg g<sup>-1</sup>, Sauer et al, 2021), surface water (0.2 µg L<sup>-1</sup>, Chen et al, 2016, 0.002 µg L<sup>-1</sup>, Zheng et al,  
358 2019; 0-0.075 µg L<sup>-1</sup>, Sauer et al, 2021), marine fish (0.005 µg g<sup>-1</sup> ww, 0.001 µg g<sup>-1</sup> dw, Akhbarizadeh  
359 et al, 2020), in bivalves, holothurians and fish in the Cabrera archipelago, not far from our sampling  
360 area (0.29, 0.1 and 0.1 µg g<sup>-1</sup> ww, respectively, Rios-Fuster et al, 2022) and in the cetacean species  
361 analysed by Montoto-Martinez et al (2021, Table 4). According to Danzl et al (2009), BPF in seawater  
362 is more biodegradable than BPP, which may explain its lower concentrations, while its higher  
363 frequency of detection may be justified by its wider use and applications. On the other hand, BPP is  
364 one of the few BP analogues with a log K<sub>ow</sub> value higher than 5 (Chen et al, 2016), which indicates a

365 tendency to accumulate in tissues, and thus be a liable factor contributing to the relatively higher  
366 concentration found in the two samples where it was detected.

367 The scarcity of literature regarding environmental concentrations of BP compounds, especially in the  
368 Mediterranean waters, as well as the almost totally lacking data on their concentration in marine  
369 wildlife, and particularly cetaceans, prevents any comparison or further speculation on the  
370 environmental factors determining the concentrations detected. Furthermore, the variety of tissues  
371 analysed within the available studies on marine mammals, only allows a qualitative comparison  
372 between the concentrations detected in the different cetacean species or geographical regions.  
373 However, even at low concentrations, the presence of several BP compounds in often a large proportion  
374 of the individuals analysed indicates that Mediterranean dolphins are somehow exposed to these  
375 contaminants. Some of the BPs detected in the muscle of striped dolphins were also detected in the  
376 Mediterranean waters, and bivalve, holothurians, and fish (Errico et al, 2017; Ozhan and Kocaman,  
377 2019; Rios-Fuster et al, 2022) from the NW Mediterranean Sea. Interestingly, the concentrations found  
378 by these authors were generally higher than those found here. The scattered nature of these data and  
379 the different measurement methods applied prevent from assessing the potential occurrence of  
380 bioconcentration or biomagnification of these pollutants. However, despite tissue levels of pollutants  
381 may vary depending on their chemical properties and the species/individual metabolism, the presence  
382 of BPs in the dolphin tissues can only be attributed to the pollution of the Mediterranean seawater,  
383 either from plastic waste, industrial and domestic effluents, landfill wastes, and/or atmospheric  
384 deposition.

#### 385 4.2 PAE concentrations

386 DEHP was the first PAE to be synthesized, in the 1930s, for its application as a plasticizer for the  
387 newly developed hard PVC (Graham, 1973). Since then, the industry of PAEs grew and diversified  
388 along with their applications, which range from home furnishings to transportation, construction,

389 apparel, food, PCP, and the medical industry (Graham, 1973). However, research demonstrated that  
390 also PAEs can leach from plastics to the environment and that the exposure to these chemicals can  
391 produce toxic effects (Warner and Flaws, 2018). Several PAE compounds, such as DEHP, DBP and  
392 DIBP, have been included as hazardous for the environment in the European REACH regulation to  
393 protect human health from chemical risks (EU 1907/2006), and their use has been banned in the EU  
394 from a number of applications (Net et al, 2015). They have also been included, along with a few others,  
395 in the list of priority hazardous pollutants in China and the USA (CPSC, 2018), and are already subject  
396 to some restrictions in the USA. Although the use of PAE has declined, they still make up over 55%  
397 of world plasticizers, which are consumed at a growing rate of 3.5% per year (IHSMARKIT, 2020) and  
398 are thus expected to end up in large quantities in the marine environment.

399 The analysis of PAEs is challenging, because the sample manipulation and the use of organic solvent  
400 may lead to possible contamination of the samples and/or of the laboratory environment (Panio et al,  
401 2020). In this study, we detected 6 of the 13 PAEs analysed, which in decreasing order of frequency,  
402 were: DEP, DBP, DEHP, DMP, DCHP, DNOP, and, in decreasing order of concentration: DBP, DEHP,  
403 DEP, DCHP, DNOP, DMP. Marine mammals are exposed to PAEs mainly through the ingestion of  
404 materials that may release these compounds once they are made bioavailable by digestive surfactants  
405 (Teuten et al, 2007), or through contaminated food, water and air. However, as PAEs are rapidly  
406 degraded in the environment (Xie et al, 2007) and metabolised by living organisms (Mackintosh et al,  
407 2004), and they do not appear to biomagnify in aquatic food webs (Mackintosh et al, 2004; Routti et  
408 al, 2021), exposure through prey ingestion is expected to be limited (Hart et al, 2020).

409 In the NW Mediterranean Sea, PAE concentrations have been found to range from 100 to 527 ng L<sup>-1</sup>  
410 in seawater, from 12 to 610 ng g<sup>-1</sup> dw in sediment, and from 0.9 to 47 µg g<sup>-1</sup> dw in zooplankton  
411 (Schmidt et al, 2021). Consistently with Garcia-Garin et al (2022) and Montoto-Martínez et al (2022),  
412 the only two studies assessing PAEs in cetacean muscle, our results showed a high variability of the

413 overall individual PAE concentrations, and identified DEHP, DEP and DBP as the most common and  
414 most concentrated compounds. Indeed, these compounds are among the most frequently detected PAEs  
415 in surface waters (Net et al, 2015; Hart et al, 2020).

416 DEHP has been detected in the Mediterranean waters of the Gulf of Marseille at concentrations ranging  
417 from 15.8 to 923.8 ng L<sup>-1</sup> (Paluselli et al, 2018), and was the more abundant PAE compound observed  
418 in the surface sediments across the Gulf of Lion (Alkan et al, 2021). It was detected in the gonads of  
419 sea urchins from Sardinia (17 ng g<sup>-1</sup>, Raguso et al, 2022), in bivalves, holothurians and fish from the  
420 Balearic Islands (mean 2.58, 1.48 and 0.88 µg g<sup>-1</sup> ww, respectively, Rios-Fuster et al, 2022) and in  
421 biopsies taken from cetaceans (including two striped dolphins) in the Pelagos Sanctuary (Baini et al,  
422 2016), where high concentrations of this compound and its metabolites were also detected in marine  
423 neuston and plankton samples (Fossi et al, 2012; Baini et al, 2016). DEHP was also detected in the  
424 cetacean samples analysed by Vorkamp et al (2004), Routti et al (2021), and Montoto-Martinez et al  
425 (2021) in the North Atlantic Ocean (Table 4). As this compound is the most widely used to increase  
426 flexibility of plastics (especially PVC), representing in 2015 over 35% of the global plasticizer market  
427 (Routti et al, 2021), it is also the most studied, along with its metabolites. Given that DEHP has low  
428 solubility in water and preferentially partitions to suspended particles and sediments, it is prone to  
429 bioaccumulate in lipid-rich tissues and to biomagnify along marine food webs, the main exposure for  
430 marine mammals happens through the food (Dziobak et al, 2021; Routti et al, 2021). These combined  
431 factors may explain why this compound was detected at the highest concentrations in our samples.

432 On the other hand, studies on low molecular weight PAEs, such as DBP and DEP, in the tissues of  
433 aquatic organisms are scarcer. Indeed, these compounds have been reported to accumulate at low level  
434 in aquatic species (Net et al, 2015), and their half-life is short both in the marine environment (*i.e.*,  
435 over 90% degradation within a week, Staples et al, 1997) and in fish tissues (Net et al, 2015). DEP was  
436 detected in the waters of Marseille Bay at concentrations ranging from 3.3 to 50 ng L<sup>-1</sup> (Paluselli et al,

2018), in a small proportion of sediment samples from the Gulf of Lion (Alkan et al, 2021), in bivalves, holothurians and fish from the Balearic Islands (0.54, 0.49 and 0.17  $\mu\text{g g}^{-1}$  ww, respectively, Rios-Fuster et al, 2022), in 40% of the samples of cetacean muscle analysed by Montoto-Martínez et al (2021), as well as in the tissues of the dolphins analysed by Page-Karijan et al (2020) and in the muscle of the fin whales analysed by Garcia-Garin et al (2022). DBP was detected in the gonads of sea urchins from Sardinia (10  $\text{ng g}^{-1}$ , Raguso et al, 2022), in bivalves, holothurians and fish from the Balearic Islands (0.78, 1.24, 0.72  $\mu\text{g g}^{-1}$  ww, respectively, Rios-Fuster et al, 2022), and in the samples of cetacean muscle analysed by Montoto-Martínez et al (2021) and Garcia-Garin et al (2022) (Table 4). The concentrations found in these studies were of comparable order of magnitude as those we detected, despite not all the cited studies analysed the same tissues, which prevents a rigorous comparison of concentrations, and the frequency of detection varied among studies and species, probably indicating that, while the metabolism of this compound is similar among cetacean species, its level of exposure varies between geographical areas. Its metabolite, MEP, was also detected in the samples of dolphin urine analysed by Hart et al (2018, 2020) and in the liver of harbour porpoises (Rian et al, 2020; Table 4).

DMP is also a low molecular weight PAE, which is used, along with DBP and DEP, as a component of industrial solvents, adhesives, waxes, inks, insecticides, PCPs and pharmaceutical products (Net et al, 2015). This compound was detected in the waters of the Marseille Bay at concentrations ranging between 0.8 and 11.9  $\text{ng L}^{-1}$  (Paluselli et al, 2018) and in the sediments of the Gulf of Lion (Alkan et al, 2021). We detected this compound in half of our samples, at very low concentrations, similar to those detected in the liver of ringed seals and polar bears by Vorkamp et al (2004), and in the muscle of fin whales by Garcia-Garin et al (2022), who observed it in a higher proportion of samples (Table 4). Rian et al (2021) also detected the presence of its metabolite, mMP, in the liver of 69% of the 100 harbour porpoises they analysed. The apparent lower prevalence of this compound in our and other Mediterranean samples, as compared to those from other areas, is likely related to different rates of

462 DMP deposition in the Mediterranean Sea than in the Atlantic Ocean, which could be the result of  
463 lower rates of production and use in the countries bordering the Mediterranean Sea.

464 Paluselli et al (2018) highlighted that the origin and aging of plastic materials may affect the  
465 concentration of PAEs in both the seawater and the guts of marine organisms. Moreover, while  
466 compounds like DEHP are denser than seawater and thus subject to sinking, phthalates can also leach  
467 from positively buoyant polymers such as polyethylene (PE), polypropylene (PP), and polystyrene  
468 (PS) (Paluselli et al, 2019). Thus, the highly variable concentrations detected, and the variability found  
469 among studies may not only derive from different environmental deposition rates among compounds,  
470 but also from different feeding habits of the cetacean species or of the individuals analysed (*i.e.*, pelagic  
471 vs benthic), along with the intrinsic differences related to the assessment of these pollutants in different  
472 tissues.

473 Finally, DCHP and DNOP were only detected in 4 and 2 dolphins analysed, respectively, at lower  
474 concentrations than most other compounds. To our knowledge, the only study reporting these  
475 compounds in a cetacean tissue is that from Garcia-Garin et al (2022), which detected DCHP in one  
476 of the fin whale muscle samples analysed, while this compound was below the limit of quantifications  
477 in the marine mammal samples analysed by Routti et al (2020) (Table 4). This result might indicate  
478 that the exposure to these contaminants in the marine environment is relatively low, but the scarce  
479 literature available prevents from relating the levels found to the local uses and production rates.

#### 480 4.3 Influence of biological traits

##### 481 *Sex*

482 None of the single BP or PAE compounds analysed was detected at significantly different  
483 concentrations in samples from adult males and females. This result is consistent with the few  
484 publications analysing these compounds in mammals of different sex, including marine mammals and

485 clinical studies in humans. Published research is consistent in stating that these compounds do not  
486 bioaccumulate in tissues; on the contrary, they are largely excreted through the urine, faeces, bile, hair  
487 or fur (Kang et al, 2006; Chen et al, 2016; Nehring et al, 2017). Dziobak et al (2021) and Rian et al  
488 (2020) also showed that the concentrations of phthalate metabolites in the urine and liver of odontocete  
489 species do not differ between sexes. However, the metabolism of each compound may vary according  
490 to its chemical characteristics, as well as to individual variables and habits like the diet (Akhbarizadeh  
491 et al, 2020). In humans, diet, lifestyle, and other individual habits, were proven to be the main factors  
492 determining different metabolite concentrations in males and females (*e.g.*, Hartmann et al, 2015;  
493 Huang et al, 2015; Silva et al, 2004). Moreover, the metabolism of female mammals often differs from  
494 that of males as a consequence of their hormone cycle, pregnancy, and lactation, which can affect the  
495 concentration of contaminants in their tissues (Aguilar et al, 1999). Indeed, laboratory studies show  
496 that female rats metabolize BPA faster than males, and different BPA concentrations have been found  
497 in the human serum, as a consequence of differences in the androgen-related metabolism (Kang et al,  
498 2007). These factors may also explain why, although the concentration of none of the compounds was  
499 significantly different between sexes, the summation of all PAE compound concentrations was overall  
500 higher in samples from males than in those from female dolphins. However, given the limited number  
501 of samples used to perform this analysis, this result is to be interpreted with caution.

#### 502 *Maturation stage*

503 In contrast with certain organic compounds such as PCBs and chlorinated pesticides that show age-  
504 related accumulation in cetaceans (*e.g.*, Tanabe et al, 1987), neither BPs nor PAEs tend to  
505 bioaccumulate in animal tissues. Indeed, most of the few studies that assessed potential variations of  
506 these compounds with the size or age of marine mammals did not detect any specific trend (*e.g.*,  
507 Garcia-Garin et al, 2022, Montoto-Martinez et al, 2021). Our results showed no significant difference  
508 between sexually mature and immature dolphins, with the exception of DMP, whose concentrations

509 were higher in samples from immature individuals. Indeed, out of the 15 samples analysed from the  
510 2014-2018 period, this compound was only detected in three immature females, while its concentration  
511 was below the LOQ in all the remaining samples from this period. Although the statistical significance  
512 of the test is likely weak due to the limited number of samples, other authors have shown that adult  
513 mammals tend to have a more efficient metabolism of some contaminants than immature specimens.  
514 Indeed, Rian et al (2020) found that the concentration of a common metabolite of PAE, the phthalic  
515 acid (PA), decreased with body length in dolphins, and Hart et al (2021) found that mean  
516 concentrations of the overall PAE analysed and of MEHP were higher in calves than in adult dolphins,  
517 possibly reflecting a more efficient biotransformation process of these compounds in adult individuals.  
518 Maternal transfer during pregnancy and through lactation may also increase pollutant concentrations  
519 in newborn or calf mammals (*e.g.*, Borrell et al 1995; Addison and Brodie, 1977; Borrell and Aguilar  
520 2005; Fouin et al, 2012; Barbosa et al, 2018); however, this phenomenon has only been reported for  
521 human milk and the more lipophilic phthalate diesters and metabolites, such as DEHP/MEHP, while  
522 information on this regard for marine mammals is absent. As both BPs and PAEs are rapidly  
523 metabolized and do not bioaccumulate in tissues, we believe unlikely that this process is relevant to  
524 explain our results. Rather, the exposure to chemicals between adult and immature dolphins may be  
525 influenced by dissimilarities in their diet, physiology and/or activity levels, as suggested by Hart et al  
526 (2018) and Rian et al (2020).

#### 527 4.3 Influence of year of death

528 The increased demand of plastic, and plastic-related materials, has led the production of plastic  
529 additives such as BP and PAEs to raise exponentially during the past decades (Net et al, 2015). Indeed,  
530 PAEs production reached 8 million tons in 2011, and in 2020 PAEs made up 55% of the world  
531 consumption of plasticizers (IHSMARKIT, 2021). Given such a wider production, a higher amount of  
532 these compounds is expected to reach the seas, either directly from spills during the production process,

533 or in the form of leachates dispersed from the large quantities of plastic or plastic-related items that  
534 increasingly pollute the seas (Jambeck et al., 2015). However, the high liability of these compounds  
535 limits their environmental persistence, and, indeed, according to several authors, atmospheric PAE  
536 concentration in the North Atlantic did not change substantially from the 1970s (Giam et al, 1978;  
537 Atlas and Giam, 1981; Bohlin-Nizzetto et al, 2018). Consistently, Garcia-Garin et al (2022) did not  
538 detect any significant temporal trend between 1986 and 2015 in the concentration of PAE compounds  
539 in samples of fin whale muscle.

540 Although our temporal analyses were based only on 24 samples and thus the observed trends must be  
541 interpreted with caution, in line with the above findings, the concentrations of most compounds did  
542 not vary significantly during the three periods (1990, 2007-2008 and 2014-2018), with the exception  
543 of two BP and two PAE compounds. Interestingly, the concentration of three (BPE, DEP, DMP)  
544 compounds showed a significant decreasing temporal trend, while concentrations of BPFL showed a  
545 significant increase in the most recent samples. Indeed, the concentrations of BPFL were higher than  
546 the LOQ only in one sample from the 90s, and in two samples from the 2007-2008 period, while they  
547 were above the limit in most of samples from the 2014-2018 period. This result might be explained by  
548 the relative novelty of this compound, which only began to be produced and used massively as a  
549 replacement of BPA in the so-called “BPA-free” products (Özkan-Kotiloğlu et al, 2022), once the use  
550 of BPA as additive was banned in numerous products in several countries and in the EU in the early  
551 2010s.

552 BPE, in turn, was detected in 90% of samples, independently of the period, but its concentrations  
553 showed a slightly decreasing trend, with the lowest concentrations occurring in the most recent samples  
554 and the peak of concentrations detected in two samples from the 2007-2008 period. The slightly higher  
555 persistence of this compound (Danzl et al, 2009) likely justifies its detection in most samples, however,  
556 as little is known on its trends of production and use, we may only assume that this compound,

557 commonly used in cyanate resins (Zheng et al, 2019), has been recently replaced by other analogues  
558 and is reaching the Mediterranean waters in reducing quantities.

559 Similarly, DMP was detected, although at very low concentrations, in all samples from the 1990s, at  
560 slightly higher concentrations in only three samples from the 2007-2008 period, and again at very low  
561 concentrations in only three samples from immature individuals from the 2014-2018 period. Finally,  
562 DEP was detected in a consistent proportion of the individuals analysed from the three periods, but its  
563 concentrations were substantially and significantly lower in the samples from the 2014-2018 period  
564 than in those from the two earlier periods. Both compounds have been included in the lists of priority  
565 pollutants in USA, EU and China (Net et al, 2015), which may imply that the rates at which they are  
566 produced and used are every time more restricted. Indeed, while the market of high molecular weight  
567 PAEs has kept generally steady in Europe, that of low molecular weight PAEs such as DMP and DEP  
568 has been decreasing since the beginning of the 2000s: in 2017, low molecular weight PAEs still  
569 represented 40% of the global plasticizers market, but only 11% of the European one (IHSMARKIT,  
570 2021). The limited number of samples hampers a proper interpretation of results also in this case.  
571 However, given the low persistence of these compounds in the marine environment, in contrast to the  
572 behaviour of persistent organic pollutants that exhibit very slow decline in concentrations in cetacean  
573 tissues (Loganathan et al, 1990), the lower frequency and concentrations observed in the most recent  
574 samples might reflect a recent shift in the commercial production of these PAEs.

575 Indeed, our sampling period spans almost three decades, during which there have been significant  
576 changes in the production and use of these chemicals. However, the observed temporal variability may  
577 also be the result of the degradation of certain compounds (biodegradation or photodegradation), while  
578 the non-detection of certain other compounds could be related to the presence of possible unknown  
579 metabolites.

580 4.4 Implications for the Mediterranean striped dolphin population

581 The Mediterranean striped dolphin is currently classified by the IUCN red list under the “least concern”  
582 category (IUCN, 2019). However, this population has experienced a series of mortality events, during  
583 1990-1992, and 2006-2007, related with morbillivirus epizootics (Aguilar and Raga, 1993; Domingo  
584 et al, 1990; 1995; Keck et al, 2010), and its individuals are known to carry in their tissues extremely  
585 high levels of pollutants (Aguilar and Borrell, 1994; Aguilar, 2000). While, in principle, there should  
586 be no direct correlation between contaminant burden and morbillivirus, it might be possible that  
587 individuals suffering from heavy contamination have a weaker immune system and thus are more  
588 susceptible to contract a virus. Unfortunately, this possibility could not be addressed here; however,  
589 the concentrations we detected were relatively low and most probably did not have any health effect  
590 on the dolphins. Despite the tissue concentration of organochlorine pollutants and mercury in striped  
591 dolphins have shown decreasing trends (Aguilar and Borrell, 2005; Borrell et al, 2014), and the  
592 concentration of plastics ingested by Mediterranean striped dolphins is generally low (Novillo et al,  
593 2020), the main threat for this species in the Mediterranean is still associated with marine pollution  
594 (IUCN, 2019). Indeed, the extreme anthropization of the basin causes these animals to be constantly  
595 and increasingly exposed to a mixture of pollutants and subject to a variety of stress factors (Marsili  
596 et al, 2018; Aznar-Alemany et al, 2021). Although the detected concentrations do not appear to be  
597 concerningly high, and neither BPs nor PAEs seem to bioaccumulate in the tissues of dolphins, the  
598 long-term exposure to these chemicals, combined with other multiple stressors, may potentially  
599 produce adverse effects to long living animals such as cetaceans (Hart et al, 2020, Dziobaak et al,  
600 2022).

## 601 **Conclusions**

602 Our results provide the first assessment of a number of BP analogues and PAE compounds in the  
603 muscle of an odontocete cetacean, and indicate that Mediterranean dolphins are exposed to these  
604 pollutants. While no significant relation was detected between the concentration of these compounds

605 and the dolphins' biological traits (sex and sexual maturation), some temporal trends emerged. The  
606 limited number of samples available (n=30) allows only a partial interpretation of results, however, the  
607 observed trends might reflect a shift in the commercial production and use of some BP and PAE  
608 compounds, and the detected values contribute to enlarge the limited knowledge base on their levels  
609 in the marine fauna. Given the increasing rate of production and use of these compounds, and the  
610 scarce information regarding their presence in the marine environment, it is of utter importance to  
611 continue investigating their levels in sensitive marine fauna like cetaceans, along with their sources,  
612 intake routes, metabolism, and potential long-term effects.

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**Declaration of interests**

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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