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Inferring trophic groups of fish in the Central-East Atlantic from eco-toxicological characterization



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1 **Inferring trophic groups of fish in the Central-East Atlantic from eco-toxicological**
2 **characterization**

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25 **Abstract**

26
27 The marine organisms are exposed to great human-induced alterations due to the
28 indiscriminate discharges into the sea, which is why the study of marine pollution is of
29 great value for each ecosystem. Each organism bioaccumulates distantly the heavy metals
30 and trace elements in its organism. Because of this it is possible to classify different
31 groups of fish according to their feeding with the content of these metals. Ten fish species
32 were grouped considering their trophic level and habitat ecology (benthic predators,
33 herbivores, omnivores, pelagic predators and superpredator) and analyzed for its metal
34 content. Statistically significant differences were found among all the fish groups, with
35 the Superpredator group containing the highest concentrations in all metals, mainly Fe
36 (103.751 ± 92.151 mg/kg) and Al (28.908 ± 21.221 mg/kg). Therefore, this study highlights
37 that the selection of the species taking into account feeding and habitat partitioning must
38 be carefully considered being crucial to identify fish groups as biological indicators of
39 marine pollution.

40
41 **Keywords:** Heavy metals, trace elements, trophic level, ecosystem, feeding

42 **Introduction**

43

44 The Canary Islands are located off the northwest coast of Africa, with nearest point to the
45 continent in the east being 100 km away and the furthest point in the west being 450 km
46 away. Due to the volcanic origin, the islands are characterized by abrupt bathymetry, with
47 sharp profiles and narrow shelves, especially in the western islands, that affects
48 oceanographic conditions and the distribution of marine organisms (Landaeta et al., 2012;
49 Coca et al., 2014; García-Mederos et al., 2015). The archipelagos far from the continental
50 coasts form a nexus of union with other islands, however, little is still known about the
51 dispersion mechanisms which led to the first colonization of these archipelagos (Rundell
52 et al., 2004, Cowie and Holland, 2006).

53 In marine environments, fishing activities have been historically directed towards specific
54 target species, in many cases leading to severe population declines or even extinctions
55 that alter the equilibrium of the ecosystem (Fujiwara, 2012). Different fishing methods
56 are usually used to catch different groups of species. For example, the most common
57 fishing methods used in the Canary Archipelago are purse seine to catch small pelagic
58 fish, trammel nets or fishing traps to catch demersal species, angling fishing targeting all
59 kinds of small and medium-sized fish, trolling to catch medium or large pelagic species,
60 also targeted by angling with live-bait -such as sand-smelts, anchovies or sardines, among
61 others, etc. (Begg, Friedland and Pearce, 1999; Ziyadah and Chouikhi, 1999; Carrera and
62 Porteiro, 2003; Lleonart and Maynou, 2003; Marçalo *et al.*, 2006, 2013; Murta et al.,
63 2008a; Fujiwara, 2012; Mateu, Montero and Carrassón, 2014; García-Mederos, Tuya and
64 Tuset, 2015; Lucena Frédou *et al.*, 2016; Corral and Manrique de Lara, 2017; Afonso *et*
65 *al.*, 2018; Pascual-Fernández *et al.*, 2018; Jurado-Ruzafa *et al.*, 2019).

66 Coastal and marine areas are integral and essential ecosystems of the Earth and are critical
67 for global food security and for the economic well-being of nations, particularly in

68 developing countries. There is growing concern about the direct or indirect introduction
69 of pollutants into the marine environment. In recent years, marine ecosystems have been
70 impacted by contaminants coming from diverse human-induced sources such as
71 processing industries, agriculture, domestic waste, mining, as well as other sources of a
72 natural origin. Nearly 70% of the pollution comes from terrestrial anthropogenic
73 activities, whose domestic, industrial and agricultural waste products are eventually
74 discharged into the coastal waters, usually by underwater discharges (Topcuo, 2003,
75 Ruilian et al., 2008, Žvab Rožič et al., 2012; Fort et al., 2016; Lozano-Bilbao et al.,
76 2018a). Some pollutants constitute a serious risk for the environment, since they are
77 highly stable chemical substances which cannot be metabolized by living organisms,
78 meaning that they are resistant to biodegradation processes. Therefore, a bioaccumulation
79 phenomena and a multiplying effect on the concentration of these substances or elements
80 occurs in the trophic level network.

81 Depending on their relative levels, the presence of heavy metals in different foods can be
82 a serious health hazard. However, the macronutrient and micronutrient metals (or trace
83 elements) many of these are required by the organisms for the proper functioning of
84 biological systems. Even more, trace metals can have beneficial or harmful properties in
85 plants, animals and humans depending on their concentration (Bustamante et al., 2016a,
86 2016b, Carravieri et al., 2014, Dorta et al., 2015; Hosono et al., 2011, Kalay et al., 1999,
87 Lozano-Bilbao et al., 2018a, 2018b, 2018c, Raimundo et al., 2013, Hinkley, 2001).

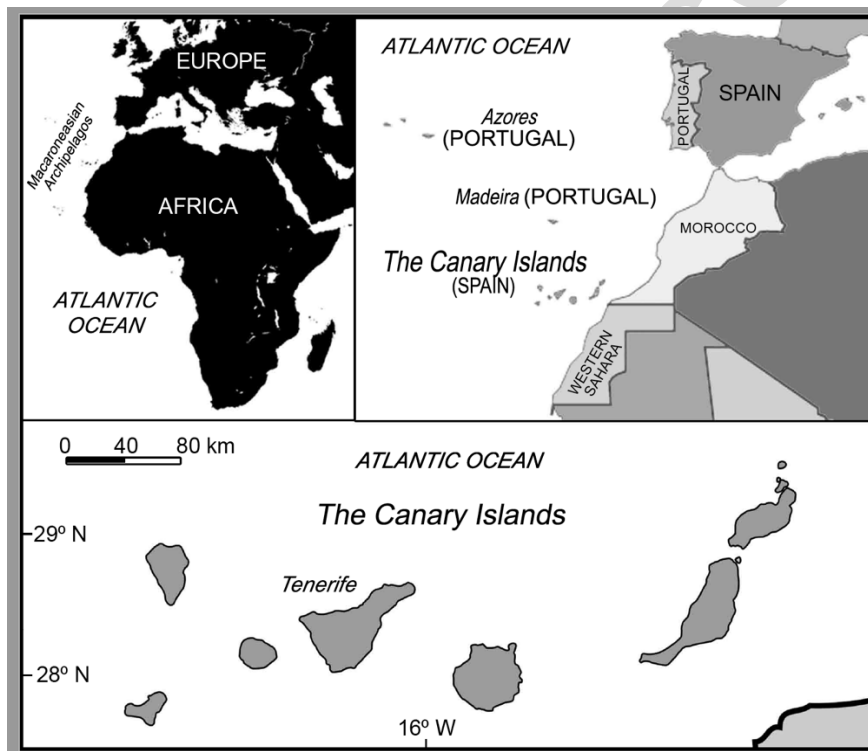
88 The main goal of the present study was to explore the relationship between the metal
89 content patterns from fish by trophic level group with ecological features, such as feeding
90 habits and habitat selection.

91

92 **Material and Methods**

93 *Biological samples*

94 A total of 352 specimens from 10 fish species were used in the study: *Thunnus*
 95 *thynnus* (Linnaeus, 1758), *Sparisoma cretense* (Linnaeus, 1758), *Chelon labrosus* (Risso,
 96 1827), *Sarpa salpa* (Linnaeus, 1758), *Diplodus sargus cadenati* de la Paz, Bauchot &
 97 Daget, 1974, *Mullus surmuletus* Linnaeus, 1758, *Serranus cabrilla* (Linnaeus, 1758),
 98 *Trachurus picturatus* (Bowdich, 1825), *Sardina pilchardus* (Walbaum, 1792) and
 99 *Scomber colias* Gmelin, 1789. All the samples were obtained from commercial landings
 100 of the small scale fishery in the Canary Islands (Fig. 1).



101

102 Figure 1: Map of the location Map of the locations of our study (Canary Islands, Spain)

103 *Sample preparation*

104 A muscle portion of approximately 5-10 g was dissected from the dorsal part of the body
 105 of each individual and preserved. The samples were dried in an oven at a temperature of
 106 70 °C for 24 hours. They were then placed in a muffle furnace for 48 hours at 450±25°C,
 107 until white ashes were obtained. If after this time the total mineralization of the samples
 108 had not been achieved, 65% HNO₃ was added to them in the fume hood and evaporated

109 on a hot plate at 70-90 °C. Once treated, they were incinerated again in a muffle furnace
110 at 450 ± 25 °C until the white ashes were obtained.

111 The white ashes were filtered with a solution of 1.5% HNO₃, made up to 25 ml for the
112 subsequent determination of the metal content by optical atomic emission spectrometry
113 with inductively coupled plasma (ICP-OES), to determine the concentration of the toxic
114 heavy metals Al, Cd and Pb and the trace elements B, Cr, Cu, Fe, Li, Ni, V, Zn in the
115 analyzed tissues.

116 *Statistical analysis*

117 The species were grouped depending on their feeding and habitat type as follows: Benthic
118 predators: *M. surmuletus* and *S. cabrilla*; Omnivores: *S. cretense*, *D. sargus cadenati* and
119 *C. labrosus*; Herbivores: *S. salpa*; Pelagic predators: *T. picturatus*, *S. pilchardus* and *S.*
120 *colias*; Superpredator: *T. thynnus*. In order to investigate whether there were variations in
121 the content and relative composition of the heavy metals and trace metals among the
122 considered groups, a statistical analysis using a distance-based permutational multivariate
123 analysis of variance (PERMANOVA) with Euclidean distances (Anderson and Braak,
124 2003) was performed. A one-way design was used with the fixed factor "feeding" with 5
125 levels of variation, depending on the type of feeding and habitat of the species analyzed.
126 The variables included in the analysis were the concentration in mg/kg of the following
127 metals: Al, B, Cd, Cr, Cu, Fe, Li, Ni, Pb, V and Zn. Relative dissimilarities among fish
128 feeding habit groups were determined using a principal coordinate analysis (PCoA) in
129 which the metals that best explained data variability were represented as vectors.

130 Finally, metals that best explained the variability of the data were explored by means of
131 univariate assessments. One-way permutational analyses of variance with Euclidean
132 distances of raw data were performed using the same design with the "feeding" factor as
133 described above.

134 In all the analyses, 4999 permutations of exchangeable units and a posteriori pairwise
 135 comparisons were used to verify the differences between the levels of the significant
 136 factors (p -value<0.01) (Anderson, 2004). The statistical packages PRIMER 7 &
 137 PERMANOVA+v.1.0.1 were used for the statistical analyses.

138

139 Results

140 Table 1 shows the mean average concentrations of each metal for each group and their
 141 standard deviations in mg/kg wet weight.

142 *Table 1. Mean average and standard deviations of the metal content by feeding/habitat group of fish*
 143 *(mg/kg, ww)*

	<i>Benthic predators</i>	<i>Herbivores</i>	<i>Omnivores</i>	<i>Pelagic predators</i>	<i>Superpredator</i>
<i>Al</i>	2.723±2.378	2.736±2.257	3.136±4.166	5.626±3.373	28.908±21.221
<i>B</i>	0.099±0.054	0.267±0.154	0.188±0.105	0.154±0.07	0.441±0.275
<i>Cd</i>	0.006±0.005	0.003±0.006	0.002±0.003	0.081±0.157	0.054±0.037
<i>Cr</i>	0.072±0.032	0.105±0.077	0.159±0.171	0.226±0.413	0.923±1.053
<i>Cu</i>	0.770±0.286	0.498±0.303	0.546±0.347	1.223±0.541	1.919±2.151
<i>Fe</i>	4.177±2.213	5.309±2.369	5.681±4.315	11.861±6.212	103.751±92.151
<i>Li</i>	0.730±0.697	0.322±0.276	0.376±0.265	0.516±0.432	0.709±0.992
<i>Ni</i>	0.094±0.145	0.027±0.02	0.052±0.124	0.295±0.616	11.561±9.152
<i>Pb</i>	0.095±0.072	0.040±0.036	0.118±0.927	0.072±0.082	0.326±0.308
<i>V</i>	0.089±0.064	0.017±0.017	0.026±0.045	0.218±0.870	0.070±0.081
<i>Zn</i>	3.177±0.895	9.958±3.288	3.501±2.056	7.649±3.288	15.549±10.635

144

145 According to their type of feeding and habitat, the PERMANOVA revealed significant
 146 differences in the content and relative composition of heavy metals and trace metals
 147 among the groups of fish considered (Table 2).

148 *Table 2. Results of the PERMANOVA, Main test.* Results of the one-way PERMANOVA analysing the
 149 variation in the content of heavy metals and trace elements in the groups of fish with contrasting feeding
 150 habits and habitats.

Source	df	SS	MS	Pseudo-F	P(perm)
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<i>Feeding</i>	4	75295	18824	87.268	0.0002*
<i>Res</i>	347	74848	215.7		
<i>Total</i>	351	1.5014E+05			

151

**p*-value<0.01

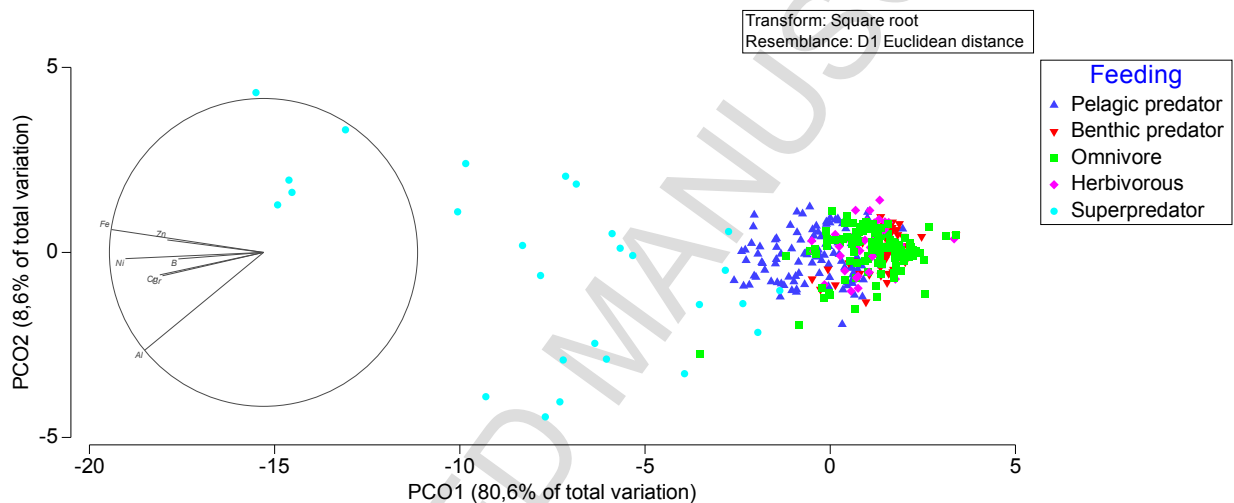
152 The *a posteriori* analyses, pairwise two by two, also showed significant differences in all
153 the analyzed groups of fish (Table 3). These results were clearly visible in the PCoA
154 analysis, which accounted for around the 89% of the total variability of the data (Fig. 2).
155 The ordination of the samples showed a clear difference of the content of heavy metals
156 in the Superpredator respect to the Benthic predators, omnivores and herbivores. These
157 three groups appeared overlapped, meaning that they have similar heavy metals content.
158 However, Pelagic predators presented a certain degree of overlap both with Superpredator
159 samples and the ‘Benthic predators-Omnivores-Herbivores’ group. Benthic predators,
160 Omnivores, appeared to have more similar heavy metals content, as can be seen by a
161 smaller segregation between points belonging to each feeding group. A higher variability
162 of data was especially observed in the case of omnivores, and samples belonging to this
163 group were more grouped together with samples belonging to Benthic predators,
164 Herbivores and, to a certain degree, with Pelagic predators. Metals that best explained the
165 variability found in the data are represented as vectors in the PCoA, which showed a clear
166 increasing pattern for Al, Fe, Ni, Cr, Cu, Zn, and B in the Superpredator group in
167 comparison to the other groups, despite the great variability among samples in this
168 feeding group.

169 *Table 3. Results of the pairwise analysis.* Results of pairwise tests examining the significant factor of
170 ‘Feeding’ obtained in the one-way PERMANOVA analysing the variation in the content of heavy metals
171 and trace elements in the groups of fish with contrasting feeding habits and habitats.

Groups	t	P(perm)
<i>Pelagic predators vs. Benthic predators</i>	7.9667	0.0002*
<i>Pelagic predators vs. Omnivores</i>	9.7478	0.0002*

<i>Pelagic predators vs. Herbivores</i>	6.1329	0.0002*
<i>Pelagic predators vs. Superpredator</i>	10.976	0.0002*
<i>Benthic predators vs. Omnivores</i>	3.415	0.0002*
<i>Benthic predators vs. Herbivores</i>	5.8315	0.0002*
<i>Benthic predators vs. Superpredator</i>	13.404	0.0002*
<i>Omnivores vs. Herbivores</i>	5.2564	0.0002*
<i>Omnivores vs. Superpredator</i>	14.365	0.0002*
<i>Herbivorous vs. Superpredator</i>	11.376	0.0002*

* p -value<0.01



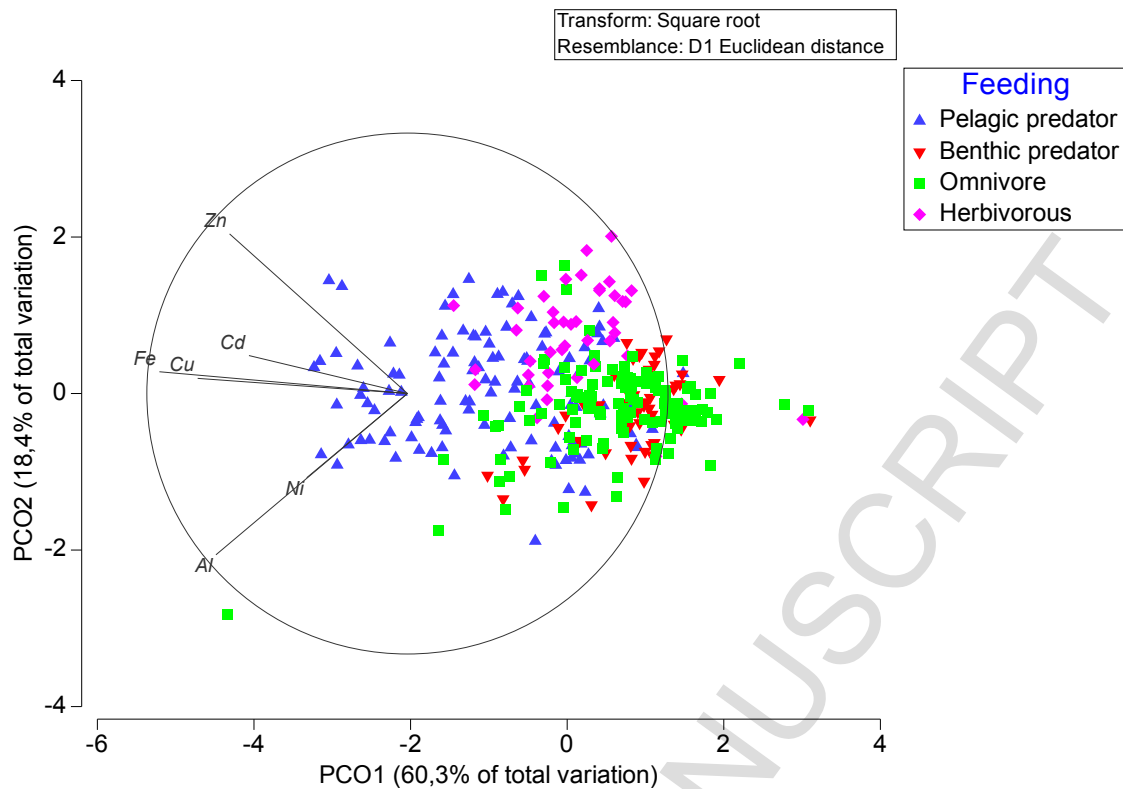
174
175

176 Figure 2. Principal coordinate analysis (PCoA) showing the first two axes (89.2% of variability), based on
177 Euclidean distances of square-root-transformed data of heavy metal and trace element content in the groups
178 of fish with contrasting feeding habits and habitats.

179

180 A second PCoA, excluding samples belonging to the Superpredator group was performed
181 to better investigate the differences among the other feeding groups, which recorded more
182 than 78% of the total data variability (Fig. 3). Iron (Fe), Cu, and Cd contents resulted
183 higher in many of the samples of the Pelagic predators.

184



185

186 Figure 3. Principal coordinate analysis (PCoA) showing the first two axes (78.7% of variability), based on
 187 Euclidean distances of square-root-transformed data of the heavy metal and trace element content in the
 188 groups of fish groups contrasting feeding habits and habitats, excluding Superpredators to better see the
 189 differences between the other feeding groups.

190

191 The results of univariate permutational ANOVAs on metals that best explained data
 192 variability, such as Al, B, Cd, Cr, Cu, Fe, Ni and Zn (Fig. 2 and 3), showed a significant
 193 effect of the feeding group in all cases. Pairwise comparisons showed significant
 194 differences between the Superpredator group and the rest of the feeding groups in all the
 195 metals considered (Table 4), with the highest concentrations in the Superpredator group
 196 (Fig. 4). Herbivorous fish differed from Pelagic predators and Benthic predators in the
 197 content of most of the analysed metals, with the exception of Cr in the first comparison,
 198 and Al and Fe in the latter, in which no differences were detected (Table 4). The observed
 199 pattern of variation of such elements showed a significant increasing trend of
 200 concentrations of Al, Cd, Cu, Fe and Ni in Pelagic predators, and higher concentrations
 201 of Cd, Cr, Cu and Ni in Benthic predators, in comparison to Herbivores (Fig. 3).
 202 Herbivores showed significantly higher contents of B and Zn (Fig. 3). Pelagic and Benthic

203 predators differed in the content of all metals except in Ni, with higher concentrations in
 204 Pelagic predators (Table 4, Fig. 4). When comparing Omnivores with Pelagic and Benthic
 205 predators, differences were found in most of the studied metals except for B and Cr in the
 206 first comparison, and Al and Zn in the second one (Table 4). Concentrations of Al, Cd,
 207 Cu, Fe, Ni and Zn in Pelagic predators resulted significantly higher than in Omnivores,
 208 but concentrations of Fe and Ni were higher in the latter (Fig. 4). Higher concentrations
 209 of Cd and Cu were found in Benthic predators in comparison to Omnivores, while B, Cr,
 210 Fe and Ni showed significantly higher concentrations in Omnivores (Fig. 4). Finally, the
 211 Omnivore and Herbivore groups resulted more similar in most of the studied variables
 212 and no differences in the concentrations of Al, Cr, Cu, Fe and Ni were found (Table 4,
 213 Fig. 3). On the contrary, the obtained concentrations of B and Zn were significantly higher
 214 in Herbivores than in Omnivores, while the content of Cd was significantly higher in
 215 Omnivores (Table 4, Fig. 4).

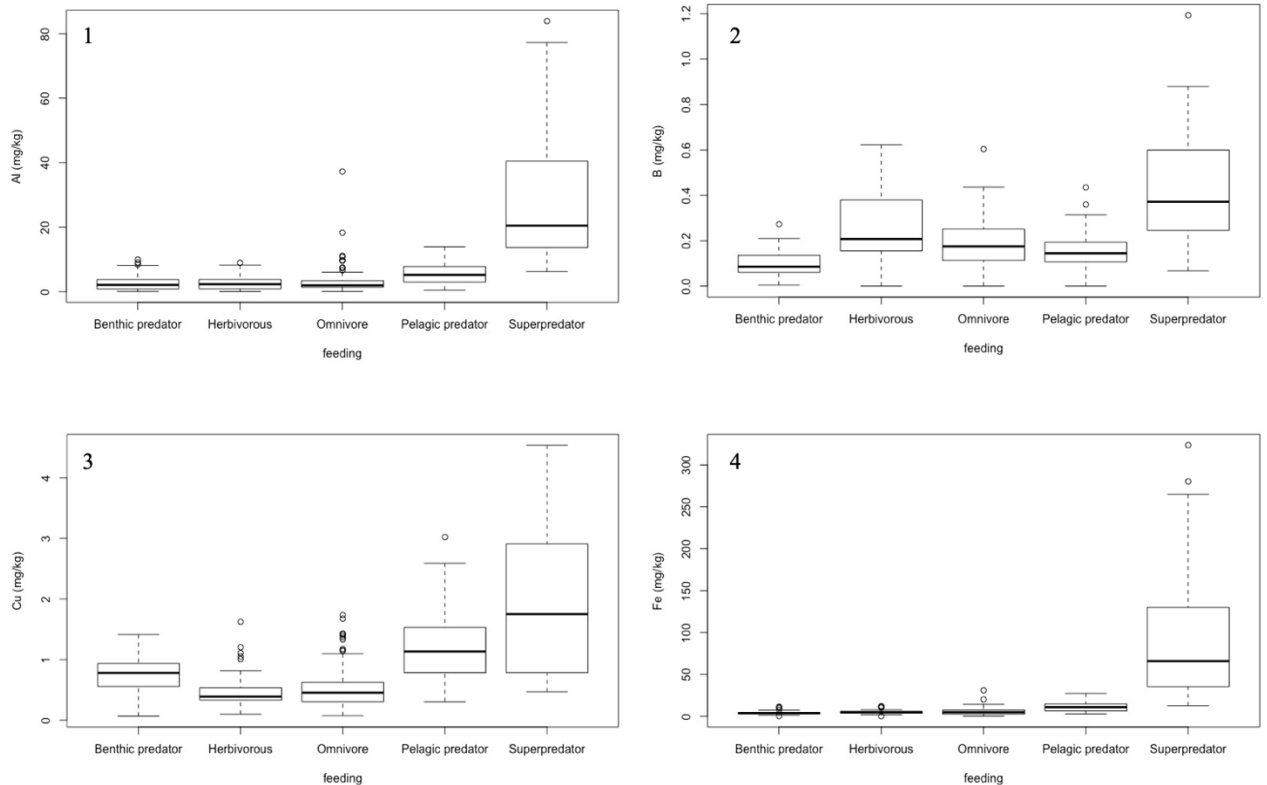
216

217 Table 4. Results of pairwise tests examining the significant factor of 'Feeding' obtained in one-way
 218 ANOVAs analysing the variation in the content of the main heavy metals and trace elements in the groups
 219 of fish with contrasting feeding habits and habitats.

Pairwise comparisons	Al	B	Cd	Cr	Cu	Fe	Ni	Zn
Pelagic predators vs. Benthic predators	0.0002*	0.0002*	0.0002*	0.0018*	0.0002*	0.0002*	0.0184	0.0002*
Pelagic predators vs. Omnivores	0.0002*	0.0254	0.0002*	0.4928	0.0002*	0.0002*	0.0002*	0.0002*
Pelagic predators vs. Herbivores	0.0002*	0.0002*	0.0002*	0.1018	0.0002*	0.0002*	0.0004*	0.0006*
Pelagic predators vs. Superpredators	0.0002*	0.0002*	0.9544	0.0002*	0.0006*	0.0002*	0.0002*	0.0002*
Benthic predators vs. Omnivores	0.537	0.0002*	0.0002*	0.0004*	0.0002*	0.0434*	0.0028*	0.384
Benthic predators vs. Herbivores	0.9776	0.0002*	0.0002*	0.0012*	0.0002*	0.017	0.0002*	0.0002*
Benthic predators vs. Superpredators	0.0002*	0.0002*	0.0002*	0.0002*	0.0002*	0.0002*	0.0002*	0.0002*
Omnivores vs.	0.5866	0.004*	0.0066*	0.063	0.4762	0.9542	0.0202	0.0002*

Herbivores								
Omnivores vs. Superpredators	0.0002*	0.0002*	0.0002*	0.0002*	0.0002*	0.0002*	0.0002*	0.0002*
Herbivores, Superpredators	0.0002*	0.0034*	0.0002*	0.0002*	0.0002*	0.0002*	0.0002*	0.0058*
* $p < 0.01$								

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Figure 4 Boxplot graphs showing mean values for the metal contents, and minimum, maximum mg/kg (ww), for each considered feeding/habitat fish groups. 1: Al; 2: B; 3: Cu; 4: Fe.

225

Discussion

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The results of the present study show the influence of ecological features in the concentration of metals accumulation in fish species, such as habitat use and feeding habits. Age, size and feeding habits of fish, as well as the amount of time spent in habitats with polluted waters during their life cycle, are known to influence metal contents in these organisms (Schuhmacher *et al.*, 1992; Kalay, Ay and Canli, 1999; Al-Yousuf, El-Shahawi and Al-Ghais, 2000; Canli and Atli, 2003). In this sense, the results show a great difference in the metal concentrations between the Superpredator group and the rest. The Atlantic bluefin tuna *T. thynnus* is the member of this group, which is the largest species

234 in the study and which has the highest trophic level (Pauly et al., 1998, Walters, Pauly
235 and Christensen, 1999). *T. thynnus* preys on many pelagic fish species, cephalopods and
236 micronekton. The Atlantic bluefin tuna have a long life and reach a large size, what leads
237 to a higher bioaccumulation of metal and trace element (Chase, 2002, Duffy et al., 2017,
238 Goldsmith, Scheld and Graves, 2017, Mariani et al., 2017; Van Beveren et al., 2017).
239 This explains the high concentrations of Al (28.908 mg/kg), Fe (103.751 mg/kg), Ni
240 (11.561 mg/kg) and Zn (15.549 mg/kg) found here. Despite the fact that other metals such
241 as Cu, Cr, B or Pb, were found in much lower levels, their concentrations in muscle of *T.*
242 *thynnus* were significantly higher than in the other fish groups. The high concentration of
243 Fe could be explained by the fact that tuna are highly migratory species with a high degree
244 of muscle irrigation which enables them to travel long distances (Di Bella et al., 2015;
245 Logan, Golet and Lutcavage, 2015; Api et al., 2018). Di Bella et al. (2015) studied the
246 concentration of heavy metals and trace elements in adult *T. thynnus* specimens in the
247 Mediterranean Sea, and found values of 18.623 mg/kg in Zn (similar to the 15.549 mg/kg
248 reported in the present study), 0.306 mg/kg of Cu (much lower than the 1.919 mg/kg
249 reported here), 0.012 mg/kg of Cd (value less than the 0.054 mg/kg found in the present
250 study) and 0.01 mg/kg in Pb (also lower than the 0.326 mg/kg of Pb found here). The
251 bioaccumulation occurring in this species is the result of their dietary exposure to metals
252 when moving around in a wide variety of environments on their migratory routes
253 (Amandè et al., 2010; Van Beveren et al., 2017). Therefore, despite the differences found,
254 it is not possible to conclude that Mediterranean tunas have a lower concentration of
255 heavy metals since this species travels thousands of kilometers during its life span
256 (Arechavala-Lopez et al., 2015; Mourente, Quintero and Cañavate, 2015; Addis et al.,
257 2016; Richardson et al., 2016)

258 The second group with higher concentration of most of the analyzed metals was the
259 Pelagic predators, which includes *S. pilchardus*, *T. picturatus* and *S. colias*, with *S.*
260 *pilchardus* being the smallest species and *S. colias* the largest, the latter even preys on the
261 other two. All these species feed on plankton, micronekton and small pelagic fish (Alonso
262 et al., 2018, Bode et al., 2018, Deudero and Alomar, 2015, Garrido et al., 2008, 2015,
263 Marçalo et al., 2006; Morote et al., 2010). The Pelagic predator group presented high
264 metal concentrations (5.626 mg/kg Al, 0.081 mg/kg Cd, 0.226 mg/kg Cr, 1.223 mg/kg
265 Cu, 11.864 mg/kg Fe, 0.218 mg/kg). The high Fe content could be due to a higher
266 musculature irrigation and, as in the Superpredator group, to the swimming capacity of
267 these species. The high Al content found both in Pelagic predators and the Superpredator
268 group seems to be a factor to be taken into account in pelagic species, which
269 bioaccumulate this non-essential metal (Cimmaruta et al., 2008, Murta et al., 2008b, Lea
270 et al., 2015; Brochier et al., 2018). Ngumbu et al. (2017) studied the metal content in *S.*
271 *colias* from Liberia, and reported values of 0.426 mg/kg for Cu, 2.503 mg/kg for Fe and
272 5.021 mg/kg for Zn. Rubio et al. (2018) studied the metal content of *T. picturatus* in the
273 Canary Islands and reported values of 1.51 mg/kg for Cu, 7.53 mg/kg for Fe and 4.69
274 mg/kg for Zn. Both studies showed lower concentrations of metals than in the present
275 study. The intrinsic variability of the pelagic environment, even within the same region,
276 as well as the numerous routes by which heavy metals can be introduced into the marine
277 environment probably exacerbate the heterogeneity of heavy metal levels in pelagic fish
278 tissues. (Canli and Atli, 2003; Licata et al., 2005; Di Bella et al., 2015).

279 The groups of Benthic predators and Omnivores had metal concentration profiles more
280 similar to each other, probably due to the fact that the members of these groups have more
281 similarities in their diet, containing different benthic components. The benthic species *S.*
282 *cretense*, *D. sargus cadenati* and *C. labrosus* were included in the Omnivore group.

283 Among the three species, *D. sargus cadenati* presents the greatest carnivorous content in
284 its diet, since it feeds on gastropods, sea urchins, amphipods, bivalves, decapods and a
285 smaller degree on algae (Sala and Ballesteros, 1997; Lloret, 2003; Clemente et al., 2010;
286 Figueiredo and Natário, 2013). *S. cretense* is a species associated with the vegetated rocky
287 seabeds of the Canary Islands, including habitats dominated by macroalgae and seagrass
288 meadows of *Cymodocea nodosa*. Its diet includes algae and various invertebrates such as
289 crustaceans, molluscs and echinoids (Board, 1956, Clemente et al., 2010, Espino et al.,
290 2015, Feline et al., 2017). In *C. labrosus*, as in the case of *S. cretense*, algae are equally
291 important in the diet and they also feed on small invertebrates such as polychaetes,
292 gastropods, bivalves and decapods (Santini et al., 2015; de Mendonça et al., 2018).
293 Abdallah (2008) studied the metal content of *S. cretense* in Egypt, and reported values of
294 0.60 mg/kg for Cd, 1.2 mg/kg for Pb and 7.4 mg/kg for Zn. Carvalho et al. (2005) studied
295 the metal content in *D. sargus cadenati* in Portugal, and found values of 0.96 mg/kg for
296 Cr and 0.01 mg/kg for Pb. Cid et al. (2001) studied the metal content in *C. labrosus* from
297 Portugal, finding values of 0.05 mg/kg for Pb, 0.006 mg/kg for Cd and 10.14 mg/kg for
298 Zn. The Cd values obtained for the Omnivore group in the present study were much lower
299 than the reported ones in the Mediterranean (Egypt), which were 0.002 mg/kg. The same
300 occurred for Pb with a value of 0.118 mg/kg, but higher than those found in Portugal.
301 Mediterranean waters is higher than in the Canary Islands, probably because this is a more
302 closed and historically more contaminated sea.

303 The Benthic predator group includes *M. surmuletus* and *S. cabrilla*. The red mullet *M.*
304 *surmuletus* inhabits muddy and sandy bottoms and the major components in its diet are
305 crustaceans such as decapods, followed by amphipods and polychaetes (Levi and
306 Francour, 2004; Labropoulou et al., 1997, Lionetto et al., 2003, Aguirre and Ciencias,
307 2005). *S. cabrilla* feeds on fish, small invertebrates such as crustaceans or mollusks and

308 also sea urchins, and it usually inhabits rocky areas and algal seabeds (Sala, 1997, Morato,
309 Santos and Andrade, 2000, Alós et al., 2011). Cid et al. (2001) studied the metal content
310 of *M. surmuletus* from Portugal and found values of 0.02 mg/kg for Cd, 0.05 mg/kg for
311 Pb and 6.67 mg/kg for Zn. Roméo et al. (1999) studied the metal content of a species of
312 the genus *Serranus* and reported values of 0.02 mg/kg for Cd and 0.3 mg/kg for Cu.
313 Bearing in mind the values of the present study, Cd was much lower than 0.006 mg/kg
314 although this was significantly higher in this group of Benthic predators than in other
315 trophic levels such as Omnivores or Herbivores. However, Cu and Pb concentrations in
316 the Canary Islands, which were 0.77 mg/kg and 0.095 mg/kg, respectively, were higher
317 than in previous studies, which may be due to the increase in pollution in coastal areas,
318 as the species of Herbivores and Omnivores analyzed here are found on the coast and this
319 makes the bioaccumulation of heavy metals and trace elements higher than that reported
320 in other studies from previous years (Verlecar et al., 2006; Vikas and Dwarakish, 2015).
321 Differences in metal concentration may be related to many aspects such as habitat, fish
322 mobility, diet, or to other characteristic behaviours of species (Henry *et al.*, 2004). The
323 species included in this trophic group, especially *M. surmuletus*, have a noteworthy
324 sediment-associated contamination due to the large amount of time spent in sediments
325 where it mainly feeds. Similarly, previous studies have found higher mean values of Cd
326 and Cu contents for flatfish also living in sedimentary sea bottoms (Zauke *et al.*, 1999;
327 Green and Knutzen, 2003).

328 Finally, the Herbivore group was represented by only one species *S. sarpa*, which is a
329 herbivorous species in its adult stage but carnivorous when young. As all the studied
330 individuals were adults, it was characterized as an herbivore feeding on green and brown
331 algae (Milazzo and Anastasi, 2006, Pagès et al., 2012, Gianni et al., 2017, 2018, Marco-
332 Méndez et al., 2017). Signa et al. (2017) studied the metal content of *S. sarpa* in the

333 Mediterranean Sea, and reported values of 0.119 mg/kg for Cd, much higher than in the
334 present study (0.003 mg/kg). In fact, the results found here on Herbivores were very
335 similar to the Omnivore group in regard to the content of most of the analyzed metals and
336 only higher levels of B and Zn were found. It is known that macroalgae bioaccumulate
337 trace metals from the surrounding water column (Roberts, Johnston and Poore, 2008; Leal
338 *et al.*, 2018). Rocky subtidal habitats of the Canary Islands are dominated by brown
339 macroalgae and other temperate algal beds, which are particularly sensitive accumulators
340 as their cell walls contain many sulphated polysaccharides for which metals display a
341 high affinity. Indeed, recent studies have reported high concentrations of Zn (250 µg/g)
342 in brown algae species, as well as Cu and Pb (Roberts, Johnston and Poore, 2008), which
343 could further explain the bioaccumulation of these metals in herbivorous fish consumers.
344 Surveys aiming to evaluate the presence of heavy metals in the marine biota, are currently
345 implemented worldwide to identify indicators of marine pollution. Certain metals, such
346 as Al, Cd, Hg and Pb are a matter of concern because of their high toxicity and tendency
347 to bioaccumulate in the food web (Lionetto *et al.*, 2003; Reguera *et al.*, 2018; Catsiki and
348 Strogyloudi, 1999). The present study has shown that in order to appropriately detect the
349 presence of such elements in marine organisms, the selection of the species must be
350 carefully considered having into account the feeding habits and habitat.

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- 639

Highlights

Ten fish species were grouped considering their trophic level

It is the top predators that have the highest metal concentration.

The groups of Benthic predators and Omnivores had metal concentration profiles more similar to each other