



# Differences in macroelements, trace elements and toxic metals between wild and captive-reared greater amberjack (*Seriola dumerili*) from the Mediterranean Sea

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## ARTICLE INFO

### Keywords:

Trace elements  
Macroelements  
Toxic metals  
Fish farming  
Greater amberjack

## ABSTRACT

Despite its legislative regulation and control, the quality and safety of aquatic products is somewhat questioned due to the potential bioaccumulation of pollutants. The elements (Al, B, Ba, Ca, Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Mo, Na, Ni, Pb, Sr, V and Zn) were determined in the liver and muscle of wild and captive-reared *Seriola dumerili* with the aim of studying possible differences between origins, and sex-related variations. Additionally, the dietary intake of these elements derived from its consumption was also evaluated. Most of the elements and metals analyzed were accumulated to a higher extent in the liver of wild specimens whereas lower differences were observed in the muscle. Overall, the elements and metal composition of wild females strongly differed from that of captive-reared specimens probably related to the mobilization of nutrients for the spawning season in wild mature females, which were greater than their captive-reared counterparts.

## 1. Introduction

The existence of metals in aquatic ecosystems may have a natural origin derived from various natural phenomena including upwelling and wind dust (Lozano-Bilbao et al., 2018) although it can also derive from anthropogenic sources. The bioaccumulation of these compounds through the trophic chain can generate risks both in the ecosystem itself and in humans that consume them (Iitembu and Richoux, 2015; Lozano-Bilbao et al., 2019a, 2020). In this sense, multiple studies have analyzed the contamination by metals of fishery products destined for human consumption while evaluating and estimating the risks derived from their intake (Afonso et al., 2017a; Herrera et al., 2020; Kalogeropoulos et al., 2012; Lozano-Bilbao et al., 2018).

Nowadays, marine fishes are one of the most important sources of food for human population due to their high nutritional value as they are rich in quality proteins and polyunsaturated fatty acids, low in saturated fats, and present good levels of fat-soluble vitamins and essential

minerals. Thus, *per capita* fish consumption has risen from 10 kg in the 1960s to 20.3 kg in 2017 (FAO, 2020). In this regard, the farming or breeding of fish in captivity is becoming more common and necessary because of the over-exploitation of most world's fishery resources. In fact, fish farming is considered the main productive activity responding to surging demand for food that is taking place due to the increase of global population (FAO, 2018). However, despite the myriad benefits of fish intake for human health and well-being (Calder, 2018; Zárate et al., 2017), the excessive consumption of large pelagic, long-lived top predator fish such as the bluefin tuna *Thunnus thynnus* and the swordfish *Xiphias gladius* might expose consumers to an excessive intake of toxic metals (Almeida et al., 2014; Bilandžić et al., 2011; Lozano-Bilbao et al., 2019a, 2020; Salam et al., 2019) and organic pollutants (Corriero et al., 2013; Desantis et al., 2005; Passantino et al., 2014). Hg, along with Cd and Pb, are non-essential heavy metals with no known essential role in living organisms that exhibit extreme toxicity even at very low (metal) exposure levels and have been regarded as the main threats to all forms

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<https://doi.org/10.1016/j.marpolbul.2021.112637>

Received 13 April 2021; Received in revised form 10 June 2021; Accepted 12 June 2021

Available online 18 June 2021

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of life including humans (Eisler, 1985; Järup, 2003; Lozano-Bilbao et al., 2020).

The greater amberjack, *Seriola dumerili*, is a large teleost fish with rapid growth, its spawning season is between May and June (Marino et al., 1995; Mylonas et al., 2004) excellent flesh quality, and high market value that is considered a leading candidate specie for aquaculture diversification (Mazzola et al., 2000; Mylonas et al., 2016), which is receiving increasing research attention in the last years (Fakriadiis et al., 2020; Jerez et al., 2018; Lazo et al., 2017; Monge-Ortiz et al., 2018; Pousis et al., 2018, 2019; Rodríguez-Barreto et al., 2014; Sarih et al., 2020; Zupa et al., 2017). Assessing the quality and safety of fishery products is essential for this industry, where comparative studies between wild and farmed individuals constitute a very active current area of research (Alexi et al., 2020; Kallio-Nyberg et al., 2015; Ruilian et al., 2008; Zhang et al., 2018). Within this context, some studies endorse that aquaculture products do not pose any more health risk than wild fish or other farmed meat products and therefore, may be considered as an excellent and secure alternative for the supply of fish today (Chanpiwat et al., 2016; Guendouzi et al., 2020; Liang et al., 2016; Squadrone et al., 2016).

The present work aims to compare the concentrations of metals and macroelements in liver and muscle of wild greater amberjack commercially captured on their spawning area in open zones of the Mediterranean Sea, with those from greater amberjack caught from the wild and reared in captivity in coastal areas. In addition, comparisons between females and males were also performed to establish possible sex-related differences. Finally, an evaluation of the dietary intake of macroelements, trace elements and toxic metals derived from the consumption of this species was also assessed.

## 2. Material and methods

### 2.1. Samples

Wild greater amberjack (11 females and 4 males) caught by a professional purse seine fishing vessel about 50 miles west of Lampedusa (Pelagie Islands, Italy) (Fig. 1) during the fishing season in 2014 and 2015 (May–June), were sampled onboard immediately after death.

The captive-reared fish (5 females, 7 males and 1 undetermined) were obtained from broodstock captured in 2011 in the area of Astakós, in the Ionian Sea (Greece) (Fig. 1) and reared for two years in the commercial farm Asterias S.A. (Greece) where they were fed fresh fish. In April 2014, the fish were transferred to Italy, placed in a 30-m<sup>3</sup> tank set with a closed recirculating system in a fish farm in Lesina (Foggia, Italy) (Fig. 1), and fed with fresh/frozen sardines and squids. Captive-reared fish died between 30 May and 01 June 2014 due to an infestation by the parasite *Amylodinium ocellatum*, being immediately sampled after death.

Samples of 5–10 g of liver and muscle from the dorsal musculature just caudal to the head were taken from each wild and captive-reared specimen and stored at −80 °C until subsequent analyses. Once in the laboratory at the University of La Laguna, samples were dried in an oven at 70 °C for 24 h and subsequently incinerated in a muffle furnace (450 °C ± 25 °C, 48 h) until obtaining white ashes. The white ashes were diluted in a 1.5% HNO<sub>3</sub> solution to a total volume of 25 mL for the determination of macroelements, trace elements and toxic metals (Al, B, Ba, Ca, Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Mo, Na, Ni, Pb, Sr, V and Zn) by Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-OES) (Elzey et al., 2012; Gutiérrez et al., 2008; Lozano-Bilbao et al., 2019b).

Several analytical procedures were taken into account during the analyses performed to control the quality, precision and data accuracy, and to ensure the comparability of the results between groups of fish

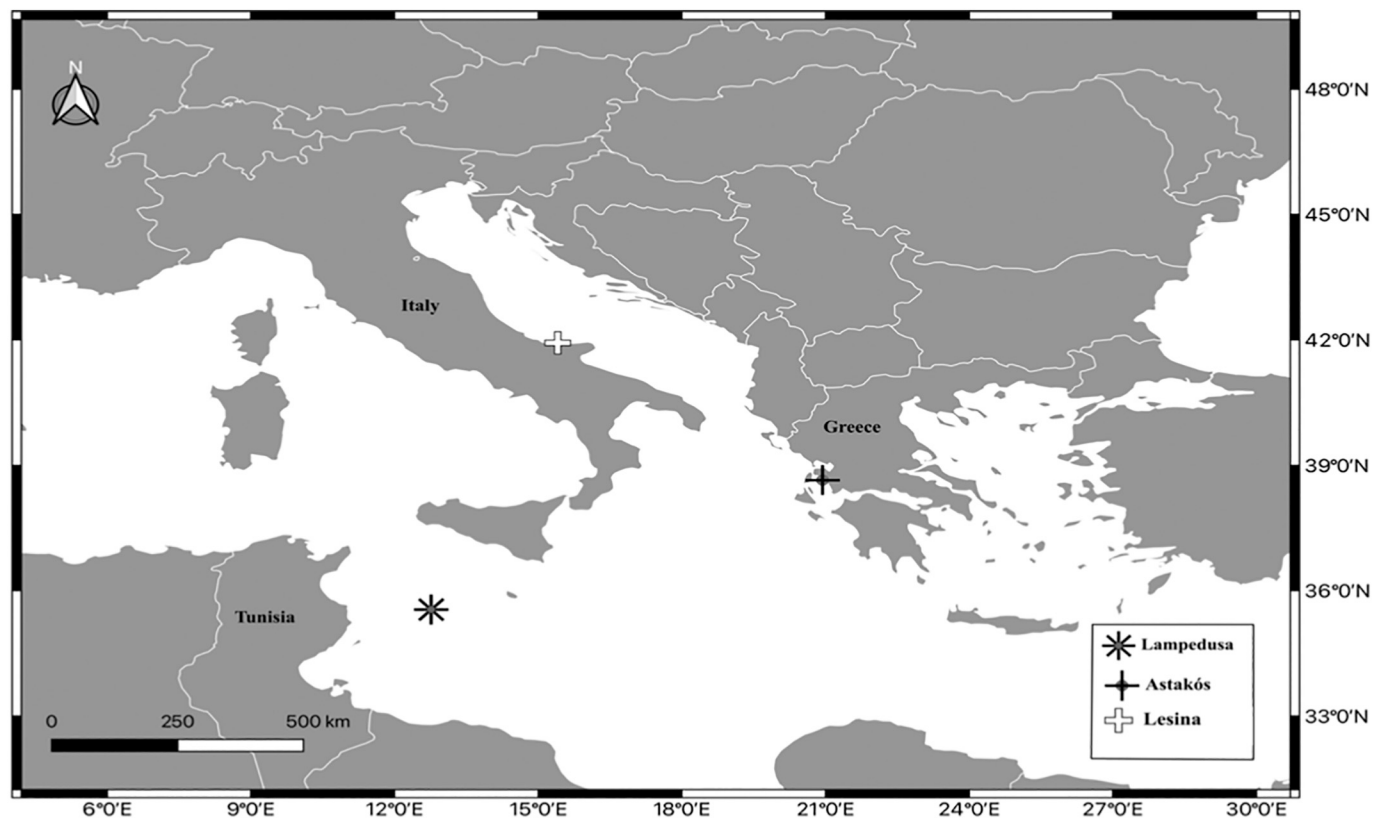


Fig. 1. Geographical locations of the capture area of wild fish (Lampedusa, Pelagie Islands, Italy) and of the two marine farms where the captive-reared fish were fattened (Astakós, Greece; Lesina, Italy).

studied. Briefly, two to three procedural blanks were prepared using the same analytical procedure and reagents, and were included within each batch of samples. Reagent blanks always had less than 1% of total contents in samples. A calibration curve was used to quantify each element, with indium (In) as internal standard (Merck, CertiPUR®). A quality control solution was used to assess the accuracy of determinations by reading every 10 samples. Moreover, certified reference materials (DORM-2, fish muscle and DORM-4, fish protein) were used to ensure precision and accuracy of results. All data collected are presented as milligrams per kilogram of tissue wet weight (mg/kg ww). The reference material was subjected to the same conditions as the samples.

## 2.2. Statistical analysis

The normality of the data was verified with the Shapiro-Wilk contrast prior to analysis. For elements and metals with a normal distribution, the independent Student's *t*-test was applied whereas the nonparametric Mann-Whitney *U* test was developed when the normality hypothesis was not fulfilled.

An analysis of covariance (ANCOVA) was performed to evaluate the effect of weight, origin, and the interaction between fixed factors (origin and sex) on toxic metals. When the hypothesis of normality and/or homoscedasticity was not verified in the proposed model, square root transformations were carried out on the response variables (metals).

The significance value in all cases was  $\alpha = 0.05$ . All statistical analyses were assessed by IBM® SPSS Statistics 20.0 software package (IBM Corp., New York, USA) for Windows.

Multivariate permutational analyses of variances (PERMANOVA) with Euclidean distances were carried out to determine the existence of significant differences in the element contents between fish groups (Anderson and Braak, 2003). In all analyses, 9999 interchangeable unit permutations and a *posteriori* comparison were used to determine the differences between the levels of significant factors (*p*-value < 0.01) (Anderson, 2004). The statistical packages PRIMER 7 and PERMANOVA+ v.1.0.1 were used to perform the analyses. A two-factor design with the factor "origin" with two levels of variation (wild and captive-reared), and the factor "sex" with two levels of variation (female and male) was used.

Relative dissimilarities among the species in each locality were determined using a principal coordinate analysis (PCoA) in which the metals that best explained data variability of trace elements were represented as vectors.

## 3. Results and discussion

### 3.1. Concentration of elements in tissues according to fish origin

The content of metals and elements (mg/kg tissue) in the liver and muscle of both wild and captive-reared greater amberjack is shown in Tables 1 and 2, respectively.

As previously reported and it is evidenced in the present work, fish metabolically active organs including gills, kidney and liver present higher concentrations of metals compared to other tissues with lower metabolic activity such as muscle (Hornung et al., 1993; Kravchenko et al., 2014). Liver is usually the storage organ for metals, where an increase in their concentration may reflect fish recent exposures. Thus, Çoğun et al. (2006), and Afonso et al. (2017a, 2017b) stated a greater accumulation of toxic metals in the liver than in the muscle of several fish species. Moreover, Al, Cd, Pb and Sr, have been reported to accumulate to a higher extent in the liver of aquatic organisms (Castro-González and Méndez-Armenta, 2008; Chae et al., 2014; Chanpiwat et al., 2016; McLaughlin et al., 1999; Wu and Sun, 2016), while in other studies, Al, Pb and Cu were more extensively accumulated in the muscle (Pethybridge et al., 2010; Squadrone et al., 2013), indicating a high interspecific variability of metals concentrations in organs under changing environmental conditions.

**Table 1**

Mean concentration  $\pm$  SD of the analyzed elements and toxic metals (mg/kg tissue) in the liver of wild and captive-reared greater amberjack (*Seriola dumerili*).

Element	Wild (n = 15)	Captive-reared (n = 14)
Al*	66.684 $\pm$ 26.777	6.535 $\pm$ 2.137
Cd*	0.469 $\pm$ 0.366	0.122 $\pm$ 0.046
Pb*	0.216 $\pm$ 0.119	0.211 $\pm$ 0.352
B	1.372 $\pm$ 0.319	1.741 $\pm$ 0.629
Ba	3.560 $\pm$ 2.215	1.255 $\pm$ 0.712
Co*	0.216 $\pm$ 0.226	0.278 $\pm$ 0.078
Cr	0.118 $\pm$ 0.091	0.155 $\pm$ 0.076
Cu*	24.769 $\pm$ 15.754	13.263 $\pm$ 4.961
Fe*	91.180 $\pm$ 46.551	263.582 $\pm$ 86.476
Li	4.146 $\pm$ 1.516	4.217 $\pm$ 1.800
Mn*	1.856 $\pm$ 0.477	1.373 $\pm$ 0.134
Mo*	0.223 $\pm$ 0.092	0.152 $\pm$ 0.039
Ni*	0.187 $\pm$ 0.060	0.119 $\pm$ 0.066
Sr*	1.275 $\pm$ 1.476	1.097 $\pm$ 0.355
V*	0.427 $\pm$ 0.352	0.356 $\pm$ 0.112
Zn*	59.715 $\pm$ 17.273	46.810 $\pm$ 10.572
Ca	158.609 $\pm$ 58.137	137.493 $\pm$ 23.424
K*	2902.819 $\pm$ 591.168	2067.987 $\pm$ 657.603
Mg	257.455 $\pm$ 45.648	230.444 $\pm$ 29.484
Na*	1431.512 $\pm$ 296.385	2327.711 $\pm$ 244.988

\* Significant differences between fish (Student's *t*-test; *p* < 0.05).

**Table 2**

Mean concentration  $\pm$  SD of the analyzed elements and toxic metals (mg/kg tissue) in the muscle of wild and captive-reared greater amberjack (*Seriola dumerili*).

Element	Wild (n = 15)	Captive-reared (n = 14)
Al*	14.524 $\pm$ 6.614	4.286 $\pm$ 1.707
Cd	0.001 $\pm$ 0.002	0.000 $\pm$ 0.001
Pb*	0.098 $\pm$ 0.036	0.039 $\pm$ 0.017
B*	0.621 $\pm$ 0.307	0.121 $\pm$ 0.044
Ba*	2.774 $\pm$ 1.661	0.643 $\pm$ 0.353
Co	0.000 $\pm$ 0E-12a	0.000 $\pm$ 0E-12a
Cr	0.099 $\pm$ 0.039	0.096 $\pm$ 0.030
Cu*	1.114 $\pm$ 0.620	1.789 $\pm$ 1.021
Fe	9.198 $\pm$ 3.921	7.379 $\pm$ 3.622
Li	1.812 $\pm$ 0.907	1.829 $\pm$ 0.851
Mn*	0.132 $\pm$ 0.041	0.106 $\pm$ 0.022
Mo*	0.021 $\pm$ 0.033	0.006 $\pm$ 0.004
Ni	0.093 $\pm$ 0.036	0.093 $\pm$ 0.091
Sr	0.577 $\pm$ 0.257	0.587 $\pm$ 0.309
V	0.117 $\pm$ 0.126	0.138 $\pm$ 0.054
Zn	6.083 $\pm$ 1.155	5.691 $\pm$ 1.574
Ca	141.284 $\pm$ 59.870	163.949 $\pm$ 42.360
K*	3125.546 $\pm$ 351.097	2653.800 $\pm$ 226.522
Mg	360.815 $\pm$ 77.423	361.320 $\pm$ 28.554
Na	909.867 $\pm$ 399.091	949.555 $\pm$ 123.835

\* Significant differences between fish (Student's *t*-test; *p* < 0.05).

In the present study, the liver of wild specimens presented higher concentrations of metals including Al, Cd, Pb, Cu, Mn, Mo, Ni, Sr, V, Zn, and macronutrients such as K, but lower amounts of Co, Fe and Na than captive-reared fish. It is noteworthy the 10-fold higher content of Al in wild specimens (Table 1). In muscle, Al, Pb, B, Ba, Mn, Mo, and K, were also greatly accumulated in wild specimens whereas Cu was more abundant in captive-reared individuals (Table 2). Overall, toxic metals accumulate to a greater extent in both liver and muscle of wild individuals, although these differences were less evident in the latter.

Wild specimens frequently develop and live in environments more exposed to contaminants than their captive-reared counterparts and fed on other wild fish that may be also contaminated (Atici et al., 2008; Kravchenko et al., 2014; Kucuksezgin et al., 2006; Lozano-Bilbao et al., 2019a; Pauly et al., 1998). Particularly, greater amberjack, like other Carangidae, aggregate around natural reefs (Heyman and Kjerfve, 2008), and also around artificial objects floating or laying on the sea

bottom such as sunken ships, oil platforms, floats or anchored devices (Castro et al., 1999, 2002). This behavior might be directly related to the higher content of most metals present in tissues of wild individual despite being captured in the open Mediterranean Sea.

Contrarily, the higher levels of Cu in the muscle of captive-reared individuals are probably related to the parasite infestation underwent by these fish which unsuccessfully received copper-based treatments a few days before their death. Hence, the concentrations obtained here must not be considered as normal values in healthy individuals.

According to the Regulation of the European Community (EC) No. 1881/2006, the maximum Pb content allowed for fish muscle is 0.30 mg/kg of fresh weight and that of Cd is 0.050 mg/kg of fresh weight. Therefore, both wild and captive-reared fish analyzed in the present study were within the permitted ranges (Table 1) and then, they were suitable for commercialization and human consumption.

### 3.2. Concentration of elements in tissues according to sex

Greater amberjack presents sexual dimorphism with females being larger at age than males (Filipovi and Raspor, 2006; Harris et al., 2007). Hence the need for characterizing and establishing the possible influence of sex in the toxicity of the edible parts of the fish. The concentration of the analyzed elements and metals in both liver and muscle by fish sex is shown in Tables 3 and 4. Regardless of the sex, metal concentrations in the liver of greater amberjack remain fairly stable (Table 3) with only differences existing in B, Fe, V, and in the macro-nutrient Na that were accumulated to a higher extent in males, while Mg was more abundant in females.

Interestingly, there was a tendency of all metals to bioaccumulate in the muscle of females although these differences were significant only for Al, Cd, Pb, B, Ba, Mg and Ni (Table 4), evidencing that larger individuals of this species concentrate elements in their edible parts.

PERMANOVA carried out in both tissues revealed that the hepatic concentration of elements and toxic metals differed between wild and captive-reared individuals in both females ( $p = 0.001$ ) and males ( $p = 0.002$ ) (Table 5). However, origin-related differences in muscle were significant only for females ( $p = 0.043$ ) probably because of nutrients mobilization for the spawning season (Albo-Puigserver et al., 2020; Jerez et al., 2006) in the larger and sexual-mature wild specimens compared to the immature captive-reared fish. Moreover, the composition of liver varied between sexes in the wild but not in captivity, probably also related to the afore-mentioned nutrient mobilization in

**Table 3**

Mean concentration  $\pm$  SD of the analyzed elements and toxic metals (mg/kg tissue) in the liver of greater amberjack (*Seriola dumerili*) according to sex.

Element	Male (n = 10)	Female (n = 15)
Al	23.605 $\pm$ 24.273	52.221 $\pm$ 40.022
Cd	0.410 $\pm$ 0.442	0.224 $\pm$ 0.111
Pb	0.252 $\pm$ 0.365	0.191 $\pm$ 0.110
B*	1.801 $\pm$ 0.657	1.355 $\pm$ 0.316
Ba	2.198 $\pm$ 1.605	2.828 $\pm$ 2.312
Co	0.318 $\pm$ 0.196	0.188 $\pm$ 0.132
Cr	0.128 $\pm$ 0.114	0.143 $\pm$ 0.057
Cu	22.602 $\pm$ 17.698	17.006 $\pm$ 7.208
Fe*	228.427 $\pm$ 117.324	118.954 $\pm$ 75.371
Li	3.816 $\pm$ 2.100	4.560 $\pm$ 1.101
Mn	1.498 $\pm$ 0.350	1.741 $\pm$ 0.480
Mo	0.203 $\pm$ 0.107	0.178 $\pm$ 0.047
Ni	0.152 $\pm$ 0.074	0.161 $\pm$ 0.070
Sr	1.357 $\pm$ 1.514	1.053 $\pm$ 0.569
V*	0.501 $\pm$ 0.335	0.291 $\pm$ 0.149
Zn	53.480 $\pm$ 15.616	54.951 $\pm$ 15.758
Ca	143.086 $\pm$ 57.267	152.152 $\pm$ 35.911
K	2287.409 $\pm$ 247.728	2709.813 $\pm$ 979.207
Mg*	221.672 $\pm$ 22.666	264.101 $\pm$ 43.986
Na*	2076.888 $\pm$ 411.883	1625.887 $\pm$ 519.353

\* Significant differences between sexes ( $p < 0.05$ ).

**Table 4**

Mean concentration  $\pm$  SD of the analyzed elements and toxic metals (mg/kg tissue) in the muscle of greater amberjack (*Seriola dumerili*) according to sex.

Element	Male (n = 10)	Female (n = 15)
Al*	6.517 $\pm$ 3.807	12.123 $\pm$ 7.872
Cd*	0.000 $\pm$ 0.000	0.001 $\pm$ 0.002
Pb*	0.051 $\pm$ 0.039	0.085 $\pm$ 0.037
B*	0.212 $\pm$ 0.161	0.512 $\pm$ 0.369
Ba*	0.927 $\pm$ 0.642	2.379 $\pm$ 1.809
Co	0.000 $\pm$ 0E-12	0.000 $\pm$ 0E-12
Cr	0.088 $\pm$ 0.024	0.105 $\pm$ 0.039
Cu	1.328 $\pm$ 0.649	1.497 $\pm$ 1.032
Fe	6.940 $\pm$ 3.470	9.427 $\pm$ 3.797
Li	1.514 $\pm$ 0.758	1.966 $\pm$ 0.915
Mn*	0.105 $\pm$ 0.024	0.132 $\pm$ 0.036
Mo	0.006 $\pm$ 0.006	0.019 $\pm$ 0.031
Ni*	0.077 $\pm$ 0.061	0.105 $\pm$ 0.069
Sr	0.545 $\pm$ 0.329	0.615 $\pm$ 0.253
V	0.104 $\pm$ 0.060	0.142 $\pm$ 0.117
Zn	5.601 $\pm$ 1.573	6.204 $\pm$ 1.122
Ca	145.118 $\pm$ 39.132	154.403 $\pm$ 61.457
K	2814.155 $\pm$ 358.618	2986.089 $\pm$ 383.807
Mg	339.210 $\pm$ 31.300	373.807 $\pm$ 69.530
Na	799.032 $\pm$ 111.117	1002.063 $\pm$ 358.173

\* Significant differences between sexes ( $p < 0.05$ ).

**Table 5**

Differences in elements and toxic metals composition of the liver and muscle of greater amberjack (*Seriola dumerili*) according to origin and sex.

Tissue	Origin	Female	Male
Liver	Wild, captive-reared	0.001*	0.002*
Muscle	Wild, captive-reared	0.043*	0.16

Tissue	Sex	Wild	Captive-reared
Liver	Male, female	0.003*	0.232
Muscle	Male, female	0.401	0.035*

\* Significant differences between groups ( $p < 0.05$ ).

wild females. Finally, no sex-related differences were found in the muscle of wild specimens ( $p = 0.401$ ; Table 5) but in captive-reared fish ( $p = 0.035$ ), due to the contents of Na and Pb (Table 6; Fig. 2).

**Table 6**

Pairwise comparison examining the significant factors 'origin and sex' obtained in ANOVAs analyzing the variation in the content of heavy metals and trace elements in the muscle of greater amberjack (*Seriola dumerili*).

Element	Wild	Captive-reared
Al	0.756	0.806
B	0.604	0.973
Ba	0.983	0.58
Cd	0.797	0.382
Co	1	1
Cr	0.253	0.1
Cu	0.114	0.432
Fe	0.845	0.213
Li	0.186	0.904
Mn	0.241	0.068
Mo	0.276	0.252
Ni	0.098	0.48
Pb	0.898	0.035*
Sr	0.651	0.934
V	0.842	0.353
Zn	0.678	0.847
Ca	0.419	0.848
K	0.886	0.592
Mg	0.612	0.099
Na	0.766	0.004*

\* Significant differences between groups ( $p < 0.05$ ).



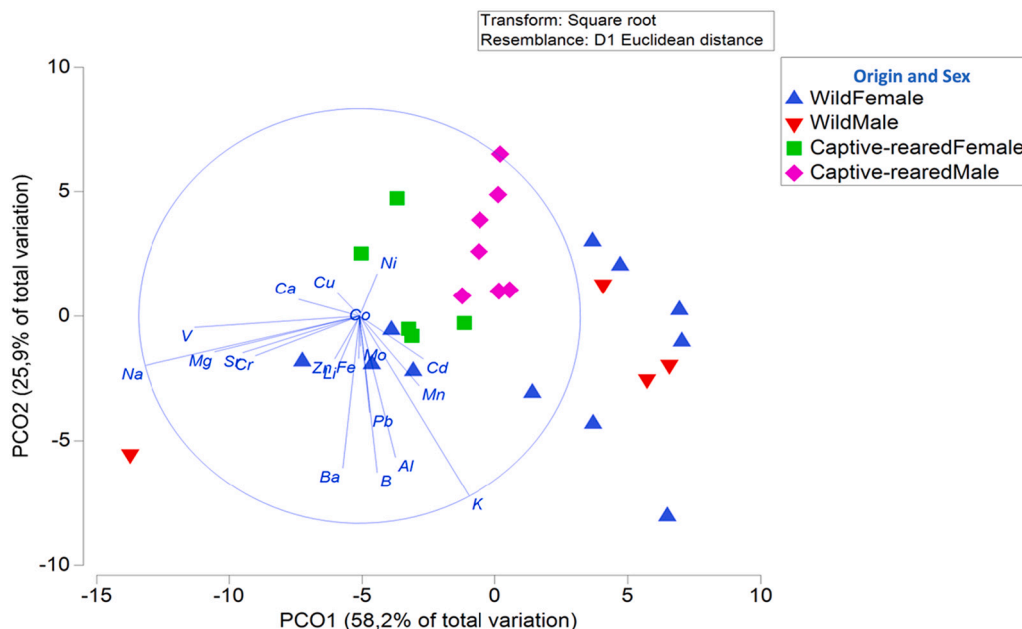


Fig. 2. Principal coordinate analysis (PCoA) showing the first two axes (84.1% of variability) based on Euclidean distances of square-root-transformed values of the heavy metals and trace elements content in the muscle of fish groups with factors (origin and sex).

### 3.3. Concentration of elements in tissues according to weight

The concentration of Pb in muscle increased with weight in wild specimens, but remained constant in captive-reared counterparts whereas Al content decreased with fish weight in wild individuals and increased in reared specimens. However, the coefficients of determination ( $R^2$ ) were very low (Table 7), thus indicating there was no correlation between the increase in concentration and fish weight for neither wild nor reared individuals.

The analysis of covariance (ANCOVA) did not show significant effects in fish muscle derived from the interaction (weight vs origin) for Al ( $p = 0.458$ ) and Pb ( $p = 0.355$ ) (data not shown). In addition, no ANCOVA was carried out for Cd as it was not present in the muscle. By contrast, there was effect of weight, origin and interaction between both factors for the hepatic concentration of Cd, which increased more sharply with weight in wild than in captive-reared fish (Fig. 3). As for muscle, there was no effect of the origin, weight, and interaction on the hepatic concentration of toxic heavy metals Al and Pb (Table 7).

### 3.4. Comparison with other studies

Although the concentrations of most elements obtained in the present study are similar to those previously reported in other species (Tables 8 and 9), some specific cases must be discussed.

Thus, the hepatic concentration of Al was much higher in wild specimens than in the captive-reared fish ( $66.684 \pm 26.777$  vs  $6.535 \pm 2.137$  mg/kg, respectively). Accordingly, Al was also abnormally high in the muscle of wild specimens (Table 6) compared to values previously reported by other authors (Çoğun et al., 2006; Lozano et al., 2009). This

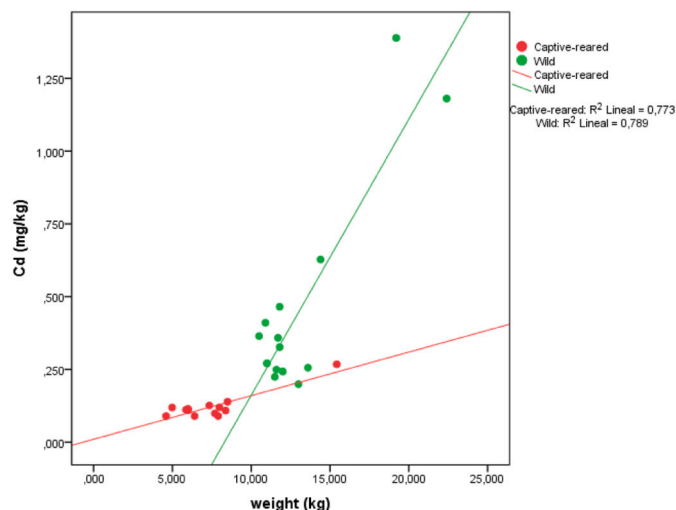


Fig. 3. Scatter plot and correlations between Cd concentration in the liver and fish weight depending on the origin; red-Captive-reared and green-Wild. The x-axis represents the weight of greater amberjack (kg) while the y-axis represents the concentration of Cd (mg/kg).  $R^2$ : coefficient of determination depending on the origin (wild vs captive-reared). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 7

Coefficients of determination ( $R^2$ ) by tissue, metal and fish origin.

Tissue	Metal	Wild	Captive-reared
Liver	Pb	0.03	0.013
	Al	0.094	0.103
	Cd	0.789	0.773
Muscle	Pb	0.120	–
	Al	0.026	0.256
	Cd	–	–

figure is extraordinarily anomalous and suggests an unusual exposure to this metal. A previous study has associated acid rain episodes in the Mediterranean Sea with the accumulation of Al in fish (Topcuoglu et al., 2003). During acid rains, the Al present in wind dust dissolves and concentrates in the water and ecosystems (Afonso et al., 2017b; Rubio et al., 2018; Topcuoglu et al., 2003). However, the precise underlying cause of the registered high levels of Al in wild fish is out of the scope of the present work. By contrast, muscle concentration of Fe was lower than that of other great predators such as the long tailed tuna (*Thunnus tonggol*) (Quratulan et al., 2015), and of deep-water fish species (Bahar et al., 2010). Zn presented similar concentrations in both groups of fish but lower values than those reported by Türkmen et al. (2008).

**Table 8**  
Summary of elements and metals in the liver of other fish species.

Element	Present study ( <i>Seriola dumerili</i> )		Others	Species	Author
	Wild (mg/kg)	Captive-reared (mg/kg)			
Al	66.684	6.535	5.19	<i>Lophius budegassa</i>	Bahar et al., 2010
Cd	0.469	0.122	0.38–1.175	<i>Thunnus thynnus</i>	Licata, 2005
			0.26	<i>Lophius budegassa</i>	Bahar et al., 2010
Pb	0.216	0.211	0.06–1.29	<i>Thunnus thynnus</i>	Licata, 2005
			1.77	<i>Lophius budegassa</i>	Bahar et al., 2010
B	1.372	1.741	1.56	<i>Chelon labrosus</i>	Afonso et al., 2017a
Ba	3.560	1.255	4.78	<i>Lophius budegassa</i>	Bahar et al., 2010
Co	0.216	0.278	0.75	<i>Lophius budegassa</i>	Bahar et al., 2010
Cr	0.118	0.155	1.14	<i>Thunnus tonggol</i>	Quratulan et al., 2015
			1.37	<i>Lophius budegassa</i>	Bahar et al., 2010
Cu	24.769	13.263	4.65–79.26	<i>Thunnus thynnus</i>	Licata, 2005
			31	<i>Lophius budegassa</i>	Bahar et al., 2010
Fe	91.180	263.582	464.36	<i>Thunnus tonggol</i>	Quratulan et al., 2015
			134	<i>Lophius budegassa</i>	Bahar et al., 2010
Li	4.146	4.217	10.2	<i>Lophius budegassa</i>	Bahar et al., 2010
Mn	1.856	1.373	0.53–1.09	<i>Thunnus thynnus</i>	Licata, 2005
			6.99	<i>Lophius budegassa</i>	Bahar et al., 2010
Ni	0.187	0.119	0.93	<i>Thunnus tonggol</i>	Quratulan et al., 2015
			1.59	<i>Lophius budegassa</i>	Bahar et al., 2010
Sr	1.275	1.097	2.03	<i>Lophius budegassa</i>	Bahar et al., 2010
V	0.427	0.356	2.30	<i>Chelon labrosus</i>	Afonso et al., 2017a
Zn	59.715	46.810	0.67–9.96	<i>Thunnus thynnus</i>	Licata, 2005
			38.2	<i>Lophius budegassa</i>	Bahar et al., 2010
Ca	158.609	137.493	213	<i>Lophius budegassa</i>	Bahar et al., 2010
K	2902.819	2067.987	2062	<i>Merluccius merluccius</i>	Bahar et al., 2010
Mg	257.455	230.444	199.32	<i>Chelon labrosus</i>	Afonso et al., 2017a
Na	1431.512	2327.711	1125	<i>Merluccius merluccius</i>	Bahar et al., 2010

*T. thynnus*, *Lophius budegassa* and *Merluccius merluccius* from Mediterranean Sea; *T. tonggol* from Arabic Sea; *Chelon labrosus* from Atlantic Ocean (Canary Islands) and *Decapterus kurroides* from Malaysia.

### 3.5. Dietary intake

The intake of elements and metals derived from the daily consumption of *S. dumerili* muscle is shown in Table 8. For each element, the established reference/recommended values for a 70 kg person have been considered, and the daily amount of greater amberjack muscle required to reach 100% of these recommendations has been estimated.

**Table 9**  
Summary of elements and metals in the muscle of other fish species.

Element	Present study ( <i>Seriola dumerili</i> )		Others	Species	Author
	Wild (mg/kg)	Captive-reared (mg/kg)			
Al	14.524	4.286	0.652	<i>Thunnus sp.</i>	Guérin et al., 2011
Cd	0.001	0.000	2.51	<i>Lophius budegassa</i>	Bahar et al., 2010
			0–0.23	<i>Thunnus thynnus</i>	
Pb	0.098	0.039	0.02	<i>Lophius budegassa</i>	
			0–0.24	<i>Thunnus thynnus</i>	Bahar et al., 2010
B	0.621	0.121	0.17	<i>Lophius budegassa</i>	Bahar et al., 2010
Ba	2.774	0.643	0.54	<i>Thunnus sp.</i>	Rainey et al., 1999
Co	0.000	0.000	0.022	<i>Thunnus sp.</i>	Guérin et al., 2011
			3.44	<i>Lophius budegassa</i>	Bahar et al., 2010
Cr	0.000	0.000	0.13	<i>Lophius budegassa</i>	Bahar et al., 2010
			0.01	<i>Thunnus thynnus</i>	Guérin et al., 2011
Cu	0.099	0.096	0.24	<i>Thunnus tonggol</i>	Quratulan et al., 2015
			0.32	<i>Lophius budegassa</i>	Bahar et al., 2010
Fe	1.114	1.789	0.13–1.66	<i>Thunnus thynnus</i>	
			12.9	<i>Lophius budegassa</i>	
Li	9.198	7.379	36.43	<i>Thunnus tonggol</i>	Quratulan et al., 2015
			9.13	<i>Thunnus sp.</i>	Guérin et al., 2011
Mn	0.132	0.106	37.6	<i>Lophius budegassa</i>	Bahar et al., 2010
			4.66	<i>Lophius budegassa</i>	Bahar et al., 2010
Ni	0.132	0.106	0.010	<i>Thunnus sp.</i>	Guérin et al., 2011
			0.02–0.30	<i>Thunnus thynnus</i>	Bahar et al., 2010
Mo	0.021	0.006	1.04	<i>Lophius budegassa</i>	Bahar et al., 2010
			0.063	<i>Thunnus sp.</i>	Guérin et al., 2011
Sr	0.093	0.093	0.011	<i>Dicentrarchus labrax</i>	Guérin et al., 2011
			0.35	<i>Thunnus tonggol</i>	Quratulan et al., 2015
V	0.577	0.587	0.54	<i>Lophius budegassa</i>	Bahar et al., 2010
			0.78	<i>Lophius budegassa</i>	Bahar et al., 2010
Zn	0.117	0.138	0.623	<i>Thunnus sp.</i>	Guérin et al., 2011
			0.018	<i>Thunnus sp.</i>	Guérin et al., 2011
Ca	141.284	163.949	1.25	<i>Merluccius merluccius</i>	Sepe et al., 2003
			13.6–68.4	<i>Thunnus thynnus</i>	Bahar et al., 2010
V	0.117	0.138	4.49	<i>Trigla gurnardus</i>	Türkmen et al., 2008
			20.8	<i>Lophius budegassa</i>	Bahar et al., 2010
Na	1431.512	2327.711	206	<i>Lophius budegassa</i>	Bahar et al., 2010
			177	<i>Micromesistius poutassou</i>	Bahar et al., 2010

(continued on next page)

Table 9 (continued)

Element	Present study ( <i>Seriola dumerili</i> )		Others	Species	Author
	Wild (mg/kg)	Captive-reared (mg/kg)			
K	3125.546	2653.800	3880	<i>Micromesistius poutassou</i>	Martínez-Valverde et al., 2000
Mg	360.815	361.320	446	<i>Micromesistius poutassou</i>	Godley et al., 1998
Na	909.867	949.555	1360	<i>Micromesistius poutassou</i>	Martínez-Valverde et al., 2000

*Thunnus* sp. and *Dicentrarchus labrax* on the French Coast; *T. thynnus*, *Lophius budegassa*, *Trigla gurnardus* and *Micromesistius poutassou* in the Mediterranean Sea; *T. thynnus* (Hellou et al., 1992) off the coast of Newfoundland; *T. tonggol* in the Arabian Sea; *Caranx crysos* off the coast of Rio de Janeiro; *Merluccius merluccius* in the Adriatic Sea.

With regard to metals with provisional tolerable weekly intake (PTWI), we consider values of amount of muscle daily needed to reach them less than a serving of 250 g, as potential toxicity. The contribution to metallic intakes derived from the muscle consumption of reared and wild specimens is quite similar (Table 8). The nutritional value of greater amberjack is not very relevant based on its contribution to the RDIs of metals. In addition, consumption of wild and cultivated specimens does not pose any risk in terms of the intake of toxic metals (Pb and Cd) since several kilos of fish should be consumed to reach the tolerable limits. However, due to the high concentration of Al in wild specimens analyzed in the present study, the consumption of a 675 g/day ration would suppose a similar dietary exposure to the PTWI of 1 kg/day set by EFSA (2008) (Table 10). Hence, it could be concluded that in our experimental specimens, the risk due to consumption of wild fish is higher than that arising from aquaculture in terms of its metallic composition. Based in our present results, it is suggested to monitoring Al contents in fishery products derived from wild greater amberjack in order to assess if the reported values are widespread in the Mediterranean population of this species in relation to its specific ecology and habits.

#### 4. Conclusions

Liver and muscle of wild greater amberjack bioaccumulated metals and macronutrients to a higher extent than those of captive-reared fish. By contrast, the content of metals in liver and muscle presented minor sex-related differences. Variations in females tissues in function of fish origin where probably related to the mobilization of nutrients for the spawning season in wild mature females which were greater than their captive-reared counterparts. There is no toxic risk for Pb and Cd derived from the consumption of wild or captive-reared fish analyzed in the present study, since it would be necessary to consume more than 1 kg per day to achieve the ADIs set for these metals. However, there could be a certain toxic risk with Al in wild individuals of the present study, given that daily intakes of 675 g of fish muscle would reach the ADI for Al. Regarding RDIs, the consumption of *S. dumerili* contributes discreetly to these nutritional intakes, so the diet should be accompanied by other foods that provide higher concentrations of these essential metals and macronutrients in order to cover 100% of the required RDI values.

#### CRedit authorship contribution statement

Sampling data and analysis were performed by all authors. ELB, NA and AJG leads the paper writing and all authors contributed to the

Table 10

Average amount of muscle from greater amberjack to be consumed to achieve daily intakes in a 70 kg person.

Element	mg/day/kg body weight	Amount of muscle needed daily to reach the RfD, LOAEL, MRL and RDI (kg/day) for a 70 kg person	
		Men	Women
PTWI	Al	0.14	0.675
	Cd	0.00036	24.990
	Pb	0.0036	2.571
	B	0.17	19.163
	Ni	0.022	16.559
RfD	Sr	0.6	72.790
LOAEL	Li	2.1	81.126
MRL	Co <sup>a</sup>	0.02	X
	V <sup>a</sup>	0.01	5.983
	Ba <sup>b</sup>	0.2	5.047
DRI	Cr	0.035	0.025
			<b>Men</b>
			24.747
			<b>Women</b>
			17.677
	Cu	1.1	69.120
	Fe	9	18
			<b>Men</b>
			68.493
			<b>Women</b>
			136.986
	Mn	2.3	1.8
			<b>Men</b>
			1219.697
			<b>Women</b>
			954.545
	Mo	0.045	150
	Zn	9.5	7
			<b>Men</b>
			109.321
			<b>Women</b>
			80.552
	Ca	900–1000	455.910–495.456
	K	3100	69.428
	Mg	350	300
			<b>Men</b>
			67.902
			<b>Women</b>
			58.202
	Na	1200–1500	92.321–115.401
			<b>Men</b>
			88.462–110.578

X: The tissue did not contain that metal; PTWI: Tolerable provisional intake per week; RfD: Estimation of the daily exposure dose that is unlikely to pose a risk to human health throughout life; LOAEL: lowest level at which adverse effects have been observed; MRL: minimum risk limit; RDI: recommended daily intake.

<sup>a</sup> An MRL has been derived for oral exposure of intermediate duration (Davis and Fields, 1958 and Fawcett et al., 1997, respectively).

<sup>b</sup> An MRL has been derived for oral exposure from chronic oral exposure (NTP, 1994).

interpretation of the results and the drafts reviews.

#### Declaration of competing interest

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

#### Acknowledgements

This work was supported by the project DIVERSIFY (KBBE-2013-07single stage, GA 603121) founded by the European Union Seventh Framework Programme. The authors are grateful to Mr Peppe, Giovanni and Vincenzo Billeci, and all the crew of the purse-seine fishing vessel 'Graziella' (Italy) for their friendly collaboration and hospitality on board during the sampling of wild greater amberjack.

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