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Baseline

Trace element concentrations in the apex predator swordfish (*Xiphias gladius*) from a Mediterranean fishery and risk assessment for consumersS. Gobert^a, V. Pasqualini^{b,c}, J. Dijoux^b, P. Lejeune^d, E.D.H. Durieux^{b,c}, M. Marengo^{a,b,*}^a Université de Liège, Centre MARE, Laboratoire d'Océanologie, Sart-Tilman, B6c, 4000 Liège, Belgique^b Université de Corse Pascal Paoli, UMR 6134 CNRS-UCPP Sciences pour l'Environnement, 20250 Corte, France^c Université de Corse Pascal Paoli, UMS 3514 CNRS-UCPP Plateforme marine Stella Mare, 20620 Biguglia, France^d Station de Recherche Sous-marines et Océanographiques (STARESO), 20260 Calvi, France

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ABSTRACT

Swordfish (*Xiphias gladius* L., 1758) is an apex predator, highly migratory meso-pelagic fish widely distributed in the Atlantic Ocean and Mediterranean. As top predators, this fish may be the end reservoir of the bioaccumulation of trace elements in a food chain because they occupy higher trophic levels and are an important food source, causing them to be potentially hazardous to consume. This study aims to investigate the concentration of 18 trace elements of Swordfish, caught in the Mediterranean Sea and to discuss human exposure risks. The mean element levels in the fish muscles were clearly below the maximum allowable concentrations established by International food safety regulations. The data suggested that the risk is minor and acceptable for human health. The findings of this study amplify the scarce database on contaminants available, especially new data on “emerging elements”, for this species from the Mediterranean Sea.

Trace elements (TE) are natural trace components of the marine environment, but their levels have increased due to domestic, industrial, mining and agricultural activities (Bakan and Büyükgüngör, 2000). TE are generally classified as essential (e.g. copper, zinc, iron, manganese), probably essential (e.g. nickel, vanadium, cobalt) and potentially toxic (e.g. arsenic, cadmium, lead, mercury) (Muñoz-Olivas and Cámara, 2001). At low levels, some TE are essential for enzymatic activity and many biological processes (Bat et al., 2012). The main roles of these essential elements can be described as functional (a catalyzing role) and structural (integrators of the organic compounds) (Mendil et al., 2010). TE become toxic when their intake is excessive or when ingested over a long time period and even potentially carcinogenic to humans (Uluozlu et al., 2007). TE can be bioaccumulated by marine organisms through a variety of pathways, including respiration, adsorption and ingestion (Türkmen et al., 2008). These elements can be biomagnified via the food chain and finally be assimilated by human marine food consumers involving health risks (Baeyens et al., 2005). The presence of trace element from anthropogenic origin in marine ecosystems has been a serious problem for the environment and human health (Araújo and Cedeño-Macias, 2016). Their intake can lead to adverse health effects like renal dysfunction, lung disease, liver failure, dysfunctions in the kidneys, chronic damage to the central and peripheral nervous system (Dadar et al., 2016). TE pollution of the

sea is less visible and direct than other types of marine pollution (e.g. Macro-waste) but its effects on marine ecosystems and humans are intense and very extensive (Erkan et al., 2009). Among the wide range of toxic substances contaminating the marine environment, a major concern has been focused on specific trace element (Castro-González and Méndez-Armenta, 2008).

Fish represents a powerful model for risk–benefit assessment (Di Bella et al., 2015). In the last years, the health benefits related with seafood consumption have been extensively publicized (Mendil et al., 2010). The world consumption of fish has increased simultaneously with the growing concern of their nutritional and therapeutic benefits (El-Moselhy et al., 2014). In addition to its important source of protein, fish typically have rich contents of essential minerals, vitamins and unsaturated fatty acids (Medeiros et al., 2012). However, the content of toxic TE in fish can counteract their beneficial effects (Castro-González and Méndez-Armenta, 2008). As recently pointed out, high concentrations of these elements are found in the Mediterranean Sea in many types of commercially important fish (Demirak et al., 2006; Kalay et al., 1999; Papetti and Rossi, 2009a).

Swordfish (*Xiphias gladius* L., 1758) is an apex predator, highly migratory meso-pelagic fish widely distributed in the Atlantic Ocean and Mediterranean (Macías et al., 2005). It is a large species of high commercial value, reaching a maximum length of 445 cm, weighing up

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to 540 kg and it can live for 25 years (Nakamura, 1985). Swordfish are opportunistic predators that feed primarily on pelagic fishes and invertebrates, particularly squid (Hernández-García, 1995). Swordfish typically forage in deep water during the day and stay in the mixed layer at night (Abascal et al., 2010). Based on stomach contents from *X. gladius*, it is most likely true that the swordfish uses its sword to kill some of its prey, as is shown by the slashes on the bodies of prey found in swordfish stomachs (B. Collette et al., 2016). Genetic studies suggest that all Mediterranean swordfish form a unique stock that is reproductively isolated from the Atlantic stock, indicating little genetic exchange occurring between the two (Kotoulas et al., 1995). Life history differences (e.g. growth and sexual maturity) have been noted between Mediterranean and Atlantic swordfish (Macías et al., 2005). In the Mediterranean a large, socio-economically important fishery targets this species (Aliçli and Oray, 2001), reported catch for 2015 was to 9.966 t (Aliçli and Oray, 2001; Neves dos Santos et al., 2016). Swordfish is a much sought-after table fish and consumer demand is increasing. The Mediterranean population of Swordfish was therefore regionally assessed as Near Threatened (NT) in an overview of the conservation status of Mediterranean fishes (Malak, 2011). This stock is not considered to be well-managed. In addition, the majority of the catch includes juveniles (below 90 cm Lower Jaw-Fork Length). This pauperization of Mediterranean swordfish population is due to a number of reasons, including overfishing and pollution (Damiano et al., 2011). Quota have now been instated for 2017 onwards in order to protect this overharvested population. The limit was set at 10,500 t for 2017 at a meeting of the International Commission for the Conservation of Atlantic Tunas (ICCAT) in Vilamoura, Portugal. It will be lowered by 3% per year from 2018 to 2022.

Studies of anthropogenic contaminants in large, long-lived predatory fish such as swordfish are important for several reasons. As top predators, this fish may be the end reservoir of the bioaccumulation of trace elements in a food chain because they occupy higher trophic levels and are an important food source, causing them to be potentially hazardous to consume (Mansour and Sidky, 2002). Trace elements accumulation in fish is dependent on numerous factors, and the accumulation pattern is the result of physiological uptake and elimination rates (Guven et al., 1999). Absorption of these elements may occur directly from the water, through the gills, but the main route seems to be the gastrointestinal absorption of those elements present in food (Olsson et al., 1998). Furthermore, because these species are apex predators, they carry out very intense metabolic activities that require a continuous supply of energy (Kojadinovic et al., 2007). As a result, their rate of predation and food consumption is extremely high, a property which contributes notably to the accumulation of pollutants (M.M. Storelli et al., 2005). As a consequence, TE concentration in the muscle of top predatory organisms, will reflect the environment in which they live (Szefer et al., 2003). Thus, fish tissues such as swordfish can be used as biomonitors for levels of TE compounds in the Mediterranean Sea.

As a result, the present study aims (i) to determine the levels of ten essential or probably essential metal elements (Co, Cr, Cu, Fe, Mn, Mo, Ni, Se, V and Zi) and the eight non-essential ones (Ag, Al, Be, Bi, Cd, Pb, Sb, Sn) in the muscle of swordfish caught in the Mediterranean Sea, (ii) to compare the relationships between elements concentrations and the correlations existing among them (iii) to estimate the weekly intake of these trace metals, comparing them with the Provisional Tolerable Weekly Intake (PTWI) (iv) to discuss human exposure risks with regards to International food safety regulations.

Thirty-three specimens of swordfish were obtained by fishing vessels from two sites (Bastia, Saint-Florent, Est and West coast, respectively) around Corsica Island (Mediterranean Sea) between December 2011 and August 2012. Samples and data were carefully collected by fishermen and scientific observers. Fishing location, length measurement to the nearest cm (Total Length - TL) and weight (kg) were recorded for each fish. A sample of around 10 g (wet weight, ww)

Table 1

Some morphometric and biological characteristics (mean \pm SD) of mediterranean swordfish, values in parentheses indicate minimum-maximum values.

Number of individual	Total length (cm)	Body weight range (kg)	Data collection (years)
33 (23 ♂, 10 ♀)	114 \pm 31 (70–160)	18 \pm 7 (4–30)	2011–2012

was then taken from the dorsal white muscle. The tissues were frozen at -20°C until analysis in the laboratory. The biological data of the specimens are shown in Table 1.

Before the analysis, samples were thawed and cleaned with ultra-pure water. Samples were mineralized in Teflon digestion vessels, in a closed microwave digestion labstation (Ethos D, Milestone Inc.), using nitric acid and hydrogen peroxide as reagents (suprapur grade, Merck). Analyses of 18 trace elements (Be, Al, V, Mn, Co, As, Se, Mo, Ag, Sn, Sb, Bi, Cr, Fe, Ni, Cu, Pb, Cd) were determined by Inductively Coupled Plasma Mass Spectrometry using Dynamic Reaction Cell technology (ICP-MS ELAN DRC II, PerkinElmer $^{\circ}$).

In order to check the purity of the chemicals used, a number of chemical blanks were run; there was no evidence of any contamination in these blanks. Analytical quality control was achieved using Certified Reference Materials (CRM), DOLT-3: dogfish liver, NIST 1566b: oyster tissue, NIST 1577c: bovine liver and NIST 2976: mussel tissue. The results obtained on the Certified Reference Materials showing good agreement with the certified values for all TE (global mean recovery was $92 \pm 16\%$), noticing that for Be and Bi no certified values were reported. For each TE, detection decision (LC), detection limit (LD) and quantification limit (LQ) were calculated, depending on their specific blank distribution (Currie, 1999). The results are expressed in milligrams of element per kilogram of body weight wet ($\text{mg kg}^{-1} \text{ ww}$).

All data were checked, beforehand, for goodness of fit to a normal distribution with Kolmogorov–Smirnov's test and homogeneity of variance using a Bartlett test. To better meet the assumptions of standard parametric statistical tests, to reduce the effect of outliers on skewing the data distribution and to bring elemental concentrations within the same range, the data were natural-log transformed. TE concentrations that were found to be below their analytical LD were considered as half of the LD value during data statistical treatment.

Multivariate analysis of variance (MANOVA) was used to test the effect of localities (Bastia/Saint-Florent), and sex (Male/Female) on eighteen element traces concentrations. Pearson rank correlations test were used to investigate the relationship between the trace metal levels between them (inter-elementary correlations) as well as the relationship with the biological data (weight). The correlation coefficient (r) was calculated together with p -values to determine the significance and strength of each correlation. A p value of < 0.05 was considered to indicate statistical significance.

Risk of TE intake on a weekly basis was estimated by calculating the respective levels found in *X. gladius*, for a person weighing 70 kg and with a weekly consumption rate of 427 g (defined for European population) (FAO, 2016). The Estimated Weekly Intake (EWI, mg kg^{-1}), was determined using the following equation:

$$EWI = (C_m * IR_w) / BW$$

C_m represents the TE concentration in fish (mg kg^{-1}), IR_w the weekly ingestion rate (kg) and BW the body weight (kg). To assess the public health risks, these weekly intake were compared with the Provisional Tolerable Weekly Intake (PTWI), recommended by the Joint FAO/WHO Expert Committee on Food Additives (JECFA). This index shows appropriate safe exposure levels and is used to estimate the amount of contaminants, ingested over a lifetime without appreciable risk (Chamannejadian et al., 2013). The European Food Safety Authority (EFSA) has established regulatory guidelines, the PTWI of Cd and Pb

Table 2

Mean (\pm standard deviation; SD) and range of trace elements concentrations ($\text{mg kg}^{-1}\text{ww}$) in the muscle of mediterranean swordfish caught in Corsica.

Element	Mean (\pm SD)	Min	Max
Ag	0.005 (\pm 0.001)	0,000	0,023
Al	0.943 (\pm 0.089)	0,311	2527
Be	0.023 (\pm 0.003)	0,000	0,070
Bi	0.000 (\pm 0.000)	0,000	0,002
Cd	0.033 (\pm 0.009)	0,003	0,290
Co	0.008 (\pm 0.003)	0,001	0,060
Cr	0.043 (\pm 0.020)	0,003	0,542
Cu	0.349 (\pm 0.029)	0,122	0,769
Fe	4.808 (\pm 0.706)	1340	18,406
Mn	0.072 (\pm 0.014)	0,018	0,374
Mo	0.009 (\pm 0.003)	0,001	0,094
Ni	0.271 (\pm 0.124)	0,003	3191
Pb	0.084 (\pm 0.066)	0,001	2196
Sb	0.008 (\pm 0.001)	0,001	0,024
Se	0.552 (\pm 0.072)	0,181	2513
Sn	0.003 (\pm 0.001)	0,000	0,026
V	0.003 (\pm 0.001)	0,000	0,017
Zn	30.275 (\pm 9.529)	4120	331,015

was 7 and 25 $\mu\text{g kg}^{-1}\text{week}^{-1}$, respectively. Therefore, PTWI of Cd and Pb for a 70 kg person is 490 and 1750 $\mu\text{g week}^{-1}$, respectively. Furthermore, a selection of essential group of TE (Cr, Cu, Fe, Ni, Zn) and their PTWI values were calculated and compared.

Trace elements concentrations (mean, standard deviation, minimum and maximum) in the muscle tissues of swordfish are shown in Table 2. The mean concentrations of trace elements are quite variable (Fig. 1) such as, Zn (121.101 ± 218.967), Al (3.770 ± 2.056) or Cr (0.172 ± 0.449). Distribution patterns in concentration of TE follows the sequence: Zn > Fe > Al > Se > Cu > Ni > Pb > Mn > Cr > Cd > Be > Mo > Co > Sb > Ag > Sn > V > Bi. Thus, among the trace elements analyzed, Zn showed the highest concentration. In contrast, concentrations of Bi, V, Sn or Ag were low. Be levels

Table 3

MANOVA results for testing the effects of location and sex on trace element concentrations in muscle of Mediterranean swordfish caught in Corsica. *f*-values and *p*-values of the tests used (**p* < 0.05; ***p* < 0.01; ****p* < 0.001).

Factor	Location		Sex	
Trace element concentrations	<i>F</i>	<i>P</i>	<i>F</i>	<i>P</i>
	2.671	0.284	2.670	0.278

were low in all individuals with a certain number of values under the detection limit of the ICP-MS. European legislation (E.U., 2008) established maximum levels for Pb and Cd, with values set at 0.3 mg kg^{-1} . For Pb and Cd the average concentrations (0.083 mg kg^{-1} and 0.033 mg kg^{-1} , respectively) observed in this study were lower than the maximum of the food safety regulations established by WHO and European Community. For Pb in most of the fish samples, values were below the guideline level (32/33 individuals) and only one tested samples exceeded this limit (Max: 2.195 mg kg^{-1}).

MANOVA results indicated that there were no significant differences in accumulation patterns of trace element between the two study areas (localities) and the males and females (sex) (Table 3).

The relationship between the fish weight and the trace element concentrations and also inter-relationship between elements in the muscles of the swordfish were investigated and shown in Table 4.

There was a significantly positive correlation between the weight of the fish and the concentrations of three trace element (Pb: $r = 0.35$, $p < 0.05$; V: $r = 0.35$, $p < 0.05$; Zn: $r = 0.60$, $p < 0.001$). For others trace elements, no significant correlations were detected between concentrations and weight of individuals.

Trace elements found in muscles also showed strong and moderate correlations with one another in some cases (see Table 4).

The PTWI values recommended by the expert committee on food additives, joint FAO/WHO Expert Committee on Food Additives (WHO, 2014) were used to compare the estimated weekly intakes of trace

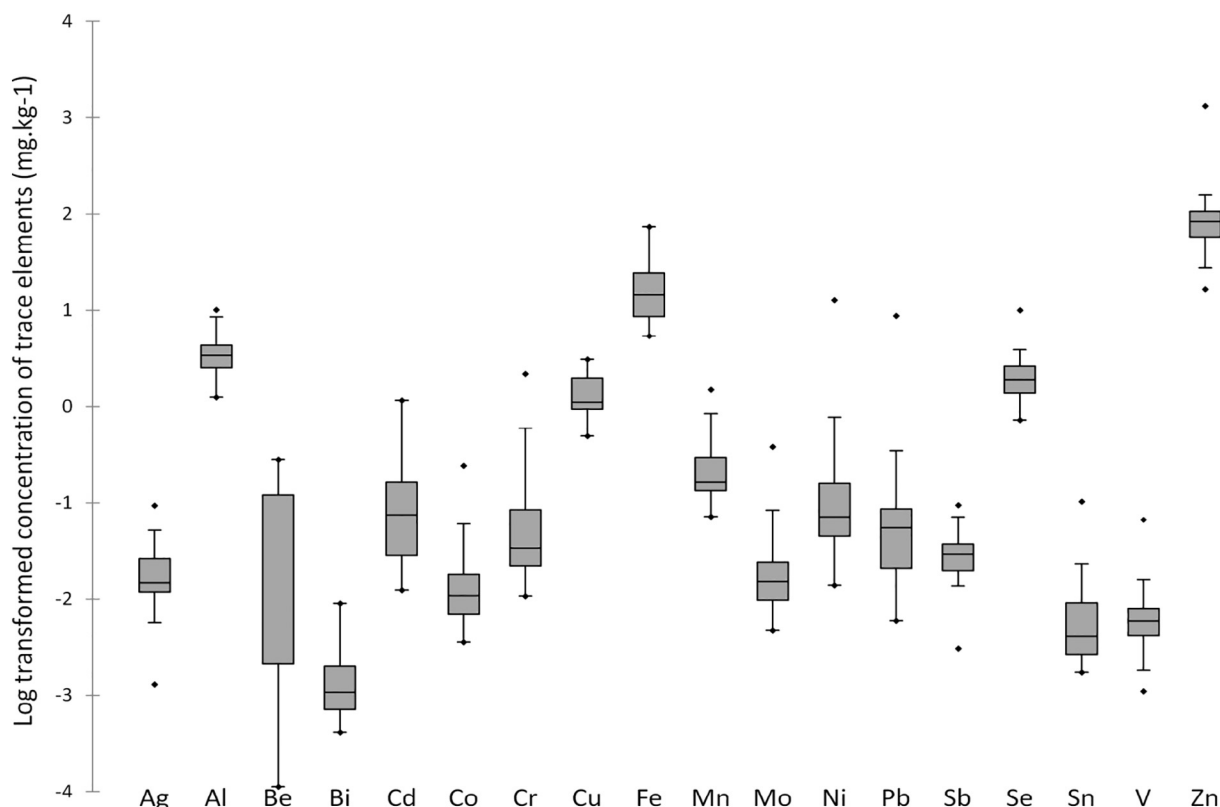


Fig. 1. Boxplot of the 18 trace elements in the muscle of mediterranean swordfish caught in Corsica.

Table 4

Pearson correlation matrix between trace element concentrations (inter-relationship) and fish weight. in the muscle of mediterranean swordfish caught in Corsica. The data in bold are statistically significant.

Element	Weight	Ag	Al	Be	Bi	Cd	Co	Cr	Cu	Fe	Mn	Mo	Ni	Pb	Sb	Se	Sn	V
Ag	0.29																	
Al	0.21	0.02																
Be	0.02	0.23	0.27															
Bi	0.14	0.15	0.45**	-0.21														
Cd	0.09	0.38*	0.09	0.21	-0.11													
Co	0.25	-0.01	0.25	-0.13	0.51**	-0.09												
Cr	0.19	-0.18	0.15	- 0.37*	0.48**	-0.16	0.73***											
Cu	0.04	-0.03	0.09	-0.23	0.34	-0.05	0.63***	0.52**										
Fe	0.20	-0.16	0.01	-0.32	0.36*	-0.09	0.56***	0.65***	0.74***									
Mn	0.15	-0.14	0.13	-0.33	0.55***	-0.29	0.82***	0.82***	0.80***	0.71***								
Mo	0.27	0.17	0.13	-0.23	0.48***	-0.08	0.80***	0.80***	0.55***	0.62***	0.78***							
Ni	0.26	0.13	0.18	-0.07	0.51***	0.01	0.85***	0.75***	0.47***	0.47***	0.76***	0.89***						
Pb	0.35*	-0.07	0.48**	-0.14	0.62***	-0.25	0.46***	0.49***	0.27	0.41***	0.54***	0.44*	0.52					
Sb	0.16	0.83***	0.16	0.37*	0.31	0.34	0.04	-0.14	-0.05	-0.22	-0.12	0.17	0.21	-0.02				
Se	0.14	-0.06	0.12	-0.13	0.26	-0.07	0.33	0.27	0.56***	0.78***	0.40*	0.34	0.17	0.23	-0.11			
Sn	0.03	- 0.42*	0.35*	-0.25	0.23	-0.24	-0.05	0.20	0.10	0.28	0.08	0.01	-0.14	0.32	-0.33	0.30		
V	0.35*	0.03	0.30	-0.05	0.37*	-0.07	0.69***	0.64***	0.57***	0.57***	0.63***	0.60***	0.51	0.46***	0.12	0.41*	0.14	
Zn	0.60***	0.10	0.25	-0.15	0.33	-0.02	0.42*	0.29	0.57***	0.58***	0.45***	0.34*	0.29	0.47***	0.04	0.59***	0.26	0.51***

* $p < 0.05$.

** $p < 0.01$.

*** $p < 0.001$.

Table 5

The estimated weekly intakes for swordfish.

Metal	PTWI ^a	PTWI ^b	EWI
Cd	7	490	0.203
Cr	637	44,590	0.262
Cu	3500	245,000	2.129
Fe	5600	392,000	29.32
Ni	35	2450	1.651
Pb	25	1750	0.511
Zn	7000	490,000	184.679

^a Provisional Permissible Tolerable Weekly Intake (PTWI) in $\mu\text{g}/\text{week}/\text{kg}$ body weight.

^b PTWI for 70 kg adult person in $\mu\text{g}/\text{week}/70$ kg body weight. EWI. Estimated Weekly intakes in $\mu\text{g}/\text{week}/70$ kg body weight.

elements in this study. The result from Table 5 suggests that the EWI of Cd, Cr, Cu, Fe, Ni, Pb and Zn by a 70 kg adult consuming 427 g of swordfish/week were all below the limit set by European regulation.

The levels of TE in swordfish were determined and assessed by comparing levels found in samples with permissible limits stipulated by various organizations. Compared to existing international food safety regulations and literature data, our results contribute to the identification of risks for mediterranean population associated with fish consumption. The European Union has established regulatory guidelines regarding dietary cadmium and lead concentrations ($0.3 \text{ mg kg}^{-1} \text{ ww}$). Cadmium accumulates in human body and may induce kidney dysfunction, skeletal damage, and reproductive deficiencies (Tuzen and Soylak, 2007). Impairment of hearing ability, anemia, renal failure, weakened immune system and premature births, are the most common symptoms of lead poisoning (Yildirim et al., 2009). The mean element levels in the fish muscles observed in this study were clearly below the maximum allowable concentrations established by WHO and European Community in food. Considering those limits, only one swordfish was reported to exceed Pb values, and all fish appeared safe for consumption regarding Cd exposure. The data modalities considered in this study suggested that the risk is minor and acceptable for human health. However, it should be taken into account that Mercury (Hg) was not measured in this study. Mercury has received much attention due to the well-known toxic effects of this metal (Storelli and Marcotrigiano, 2001). Potential dietary exposure of Hg continues to be a subject of research, regulation and debate (Storelli et al., 2007). Given its capacity for biomagnification along food webs, mercury is often present at high

levels in marine species. Several authors (Damiano et al., 2011; Papetti and Rossi, 2009b; Pastor et al., 1994; M.M. Storelli et al., 2005; Storelli and Marcotrigiano, 2001) reported moderate to high levels of mercury in swordfish caught in the Mediterranean Sea. Swordfish, usually accumulates mercury as methyl mercury, while inorganic mercury usually represents a minor proportion of the total mercury present in the tissues (Vlieg et al., 1993). Methyl mercury does not occur naturally in water, and its presence in fish muscle is due to in vivo biochemical transformation or by ingestion of preformed methyl mercury along the food chain (Mendez et al., 2001). Consequently, a large percentage of Hg is present as toxic MeHg in the edible portions of fish consumed by man (Kojadinovic et al., 2007b). From a public health point of view, the consumption of fish with high mercury content does not represent a hazard, provided that it is not eaten on a regular basis (Mendez et al., 2001). Despite the fact that our results do not show high concentrations of TE and associated risks, it remains important that higher concentrations could be found in particularly large and elderly individuals due to bioaccumulation on certain elements.

Concerning the estimation of the potential public health risks, the PTWI values were used to compare the estimated dietary intakes of trace elements in this study. The estimated weekly intake for cadmium and lead were far below the established PTWI values, it may be concluded that consumption of this species is not a problem on human health. Furthermore, the estimated intakes of chrome, copper, iron, nickel, and zinc from weekly consumption of swordfish are lower than the respective established PTWI for these elements and data modalities considered in this study.

This permissible limits should be considered as a suggested value and not an absolute one that provides a margin of safety (Onsanit et al., 2012). The regular consumption of fish is important and increasing, due to the nutritional values linked to fatty acids, some vitamins, minerals and protein, and can make a positive contribution to the prevention of cardiovascular disease and the development of the foetus (Di Bella et al., 2015). The analyzed swordfish samples represent a good nutritional source of essential trace elements. Thus, combining the detection of contaminants concentrations in food and the estimated consumption limits is of great relevance in view of assessing the balance between benefits and risks (Copat et al., 2013).

There have been very few studies on trace elements in swordfish in the Mediterranean Sea and around the world. But it should be noted that fish size, trophic position, diet and the geographical areas (both

biotic and abiotic factors) may have variable effects on the bioaccumulation of trace elements in swordfish populations. Hence caution is needed in data interpretation comparing with previous studies around the world. The comparison of our results with published data showed lower levels of lead (0.084 mg kg^{-1}) than those detected in specimens from the Mediterranean Sea (0.970 and 1.049 mg kg^{-1} , respectively) (Damiano et al., 2011; Papetti and Rossi, 2009a). Thus, in general lead levels in our study were almost twelve times lower than mean lead levels in different mediterranean swordfish (e.g. Damiano et al., 2011). But our results are consistent with other studies conducted in the Mediterranean reported by Storelli et al., 2005, and by Kojadinovic et al., 2007 around the Reunion Island (0.050 and 0.030 mg kg^{-1} , respectively).

In the same way, the range of cadmium in this study is quite different (0.033 mg kg^{-1}) from other values reported in the literature in the Mediterranean Sea (e.g. respectively 0.158 and 0.005 mg kg^{-1} , Storelli et al., 2005, Damiano et al., 2011). Thus, cadmium levels in our study were almost seven times higher than mean cadmium levels in other mediterranean swordfish (e.g. Damiano et al., 2011). However, cadmium levels observed are similar to other studies worldwide (e.g. 0.059 mg kg^{-1} , Bodin et al., 2016).

Mean values of copper found in swordfish (0.349 mg kg^{-1}) were similar to concentrations recorded in the muscle from Indian Ocean, which reached a mean of 0.342 mg kg^{-1} (Bodin et al., 2016). But these results were slightly higher than those observed in swordfish in the Indian Ocean and Atlantic Ocean (both 0.200 mg kg^{-1} , Kojadinovic et al., 2007; Olmedo et al., 2013). In comparison with data reported by other authors, mean levels of iron (4.808 mg kg^{-1}) were similar to those reported by Kojadinovic et al., 2007 and Bodin et al., 2016 in the same species (5.90 and 4.124 mg kg^{-1} , respectively). The detected chrome and selenium levels (0.043 and 0.552 mg kg^{-1} , respectively) were in good agreement with levels in swordfish in others studies (0.044 and 0.763 mg kg^{-1} respectively; Bodin et al., 2016). Our data (0.072 mg kg^{-1}) were in accordance with manganese concentrations measured in fish from the Indian Ocean (0.060 mg kg^{-1} , Kojadinovic et al., 2007), while lower values ranging from 0.013 to 0.036 mg kg^{-1} were reported in swordfish from Atlantic Ocean and Indian Ocean (Olmedo et al., 2013; Bodin et al., 2016). Zinc values in this study were high (from $30.275 \text{ mg kg}^{-1}$), exceeding by far those reported in the Atlantic Ocean and the Indian Ocean (5.637 and 7.480 mg kg^{-1} respectively (Olmedo et al., 2013; Bodin et al., 2016). But these values were comparable with those reported by Kojadinovic et al. (2007) (22.9 mg kg^{-1}). The observed variability of TE levels in the swordfish might be a result of ecological needs, metabolism, and feeding patterns (Yilmaz, 2003).

The present work is the first study to examine a large range of trace elements (18) in swordfish. As far as we know, it is the first time that these TE (Ag, Al, Be, Bi, Co, Mo, Sb, Sn, V) were measured in this species. This study completes the database and brings new knowledge concerning levels of trace elements in *X. gladius*.

The relationship between the fish weight and the TE levels in the muscles was investigated. There was a significantly positive correlation between the weight and the concentrations of three TE (Pb: $r = 0.35$, $p < 0.05$; V: $r = 0.35$, $p < 0.05$; Zn: $r = 0.60$, $p < 0.001$) for *X. gladius*. In addition to the high intake of contaminants due to the high metabolic rates of the large predators, these results reflect the biomagnification process as the contaminants move up the food chain (Damiano et al., 2011). This process occurs because consumers feeding at higher trophic levels eat larger preys with higher body burdens that smaller ones (Bodin et al., 2016). Larger individuals have shown higher TE concentrations, as was previously reported in studies in certain fish species including swordfish (Mendez et al., 2001; Storelli and Marcotrigiano, 2001).

Various degrees of correlations were found between the elements. These accumulation relationships between TE could have negative correlations where metals compete, or positive correlations where TE

accumulate together and influence one another (Renieri et al., 2014). For example, a significant positive correlation ($p < 0.001$) between Zn and Cu, Pb and Bi, Mo and Co or Fe and Cr concentration were found. A significant negative correlation ($p < 0.05$) between Ag and Sn, Cr and Be, has been found.

Associations between these elements may reflect the biochemical regulation of element concentration or a requirement of elements (such as Zn and Cu) for the synthesis of detoxifying proteins enzymes (e.g., glutathione and dismutases) as a feedback mechanism for an increase in toxic elements (Chang et al., 1998; Joyeux et al., 2004). These results can also show that some elements have similar sources (e.g. feeding) (Yilmaz et al., 2007). Thus, the correlations of TE (positive or negative) in fish tissues observed may be related to the elemental regulation which is affected by metabolic activity, environmental conditions and physiological needs (Kojadinovic et al., 2007).

Despite the human fishing pressure on swordfish, there is a great lack of knowledge on trace element levels in these fish (Kojadinovic et al., 2007). The findings of this study amplify the scarce database on contaminants available, especially new data on “emerging elements”, for this species from the Mediterranean Sea. Data on TE in swordfish have served to warn of the possible risk related to the consumption, although the data considered in the study indicate that there is no risk for a controlled ingestion.

Due to the increasing environmental pressure on the Mediterranean Sea, a regular monitoring of TE levels in marine organisms is necessary to prevent any further environmental deterioration and to assess the human exposure. These data on contaminant levels in fish from particular regions of the world could allow people to make informed decisions about which fish to eat to reduce their risk from the contaminants (Tepe, 2009).

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