Version of Record: https://www.sciencedirect.com/science/article/pii/S0048969719330487
Manuscript_f13bfe159b4ff3d5bdb3412f1fc6e904

# Title: Trace element contamination in fish impacted by bauxite red mud disposal in the Cassidaigne canyon (NW French Mediterranean)

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Abstract:

From 1966 to 2015, the Gardanne alumina refinery discharged some 20 million tons of bauxite residue (called red mud) into the Cassidaigne Canyon (northwest French Mediterranean) with impacts on local ecosystem functioning. Although these red muds contained high levels of trace elements (TE), in particular titanium (Ti), vanadium (V), aluminium (Al) and arsenic (As), surprisingly, their impacts on fish contamination levels and the risk related to fish consumption have been little studied until now. Here, 11 trace elements (Al, As, Cd, Cr, Co, Cr, Mn, Ni, Pb, Ti and V) were analyzed in muscle and, when possible, liver, from 1 308 fish of 26 species from an impacted zone in the vicinity of the Cassidaigne Canyon and a reference zone, unaffected by red mud disposals. Moreover, 66 arsenic speciation analyses were performed. Although the impact of human activities on the levels of fish contamination by trace elements is generally not easy to assess *in situ* because it is blurred by interaction with biological effects, we highlighted significant contamination of the fish species collected from the Cassidaigne Canyon, especially by the main trace elements attributable to the discharges of the Gardanne alumina refinery, namely Al, V and Ti. Moreover, inorganic toxic As concentrations were higher in the impacted zone. The results of this baseline research also confirmed the concern previously raised regarding Hg in Mediterranean organisms and that trace element contamination levels in fish are generally negatively related to fish length for all TE except Hg.

Keywords: Trace element, bioaccumulation, arsenic speciation, ICP-MS, fish, industrial discharge

# Introduction

Hazards to human health stemming from exposure to individual contaminants or groups of contaminants have emerged as a major concern in recent years (e.g. Copat et al., 2013; Damiano et al., 2011; Miniero et al., 2014; Percin et al., 2011; Suarez-Serrano et al., 2010; Turkmen et al., 2009). Among the wide range of toxic substances contaminating the marine environment, specific focus has been given to trace elements (Castro-Gonzalez and Mendez-Armenta, 2008). Naturally present in the Earth's crust, trace elements (TE) are also found in the marine environment (Bryan, 1971; Mason, 2013). However, human activities have considerably increased their spread into aquatic ecosystems (Asante et al., 2010; Nriagu and Pacyna, 1988; Squadrone et al., 2016), especially those in marine coastal areas (Ourgaud et al., 2018). TE are generally classified into two categories on the basis of their metabolic role and their regulation by organisms. Essential TE (*e.g.* iron, copper and zinc) are involved in metabolic processes, including functional and structural ones (Amiard et al., 1987). They are generally efficiently regulated by marine animals, and present a narrow range of variation in their tissues, except when ambient concentrations in seawater or food reach very high concentrations (Amiard et al., 1987; Chapman, 1996; Vallee and Auld, 1990), or when they are involved in a specific metabolic activity at a specific life stage (*e.g.* juvenile growth; Endo et al., 2008). On the contrary, some TE (*e.g.* cadmium, lead and mercury) are not essential for marine life and their concentrations in organisms depend mainly on environmental levels (Amiard et al., 1987). Whether essential or non-essential, TE become toxic above a certain threshold (Amiard et al., 1987).

From 1966 to 2015, the Gardanne alumina refinery discharged bauxite residue (called red mud) through a pipe at 320 meters depth in the Cassidaigne Canyon in the northwest French Mediterranean. This mud then spread along the canyon axis and on its lateral flanks to great depth (>2000 m). A centimetric red layer of bauxite residue can now be observed along the canyon axis, more than 50 km away from the pipe. The red mud contained high levels of TE, in particular titanium (Ti), vanadium (V), aluminium (Al), and to a lesser extent arsenic (As) (Dauvin, 2010; Fontanier et al.,

2012). These elements are present in several mineral oxi-hydroxide phases (hematite, goethite, ilmenite, rutile, anatase, brookite and kaolinite) that are not dissolved by the industrial process used to extract alumina from bauxite ore at the Gardanne alumina refinery (Fontanier et al., 2012). Red mud disposal into the Cassidaigne Canyon was expected to lower the biodiversity and abundance of marine communities and to increase the potential uptake of bioavailable TE into the tissues of marine organisms, their bioaccumulation through food webs and ultimately into human fish-consuming communities (Ellis et al., 1995; Gray et al., 2003; Metian et al., 2013; Swales et al., 1998; Williams et al., 1999).

Since the 1960s, several environmental impact studies of the Gardanne alumina production activity have been performed to document the effects of these disposals on biodiversity (Dauvin, 2010) but, surprisingly, up to now the impact of red mud residues in the Cassidaigne Canyon on TE contamination of fish and the risk related to fish consumption have been studied only twice. In 2004, 9 TE were analyzed in 28 samples from 12 fish species and in 2013, 27 TE were analyzed in 11 samples from 8 fish species (ANSES, 2016; Dauvin, 2010). These studies did not identify any cumulated risk linked with fish consumption (Dauvin, 2010). However, their conclusions were limited: (*i*) because the number of fish analyzed per species was very low; and (ii) because thresholds were not compared with baseline values measured in a control zone, precluding any clear conclusion on actual contamination. Indeed, for elements without regulatory thresholds such as Ti and V, a robust impact assessment should include a comparison of trace metal concentrations in fish from the impacted area and from an non-impacted area chosen as control. Moreover, As is one of the elements present in the red muds discharged in the Cassidaigne Canyon. Arsenic can biomagnify in food webs (Francesconi, 2010; Rahman et al., 2012), but this element exists in a number of chemical forms in nature, all with very different toxicity characteristics, with inorganic species being the most hazardous (Saha et al., 1999). Therefore, the determination of the total As content is certainly not sufficient for estimating its potential toxic impact and its speciation must be studied. Indeed, very few data are available on As speciation in the Mediterranean context (see Fattorini et al., 2004;

Storelli et al., 2005; Storelli and Marcotrigiano, 2000). To our knowledge, As speciation in fish collected close to the Cassidaigne Canyon has never been studied.

In this context, the specific aim of this study was to assess, for the first time, the direct impact of red mud disposal in the Cassidaigne Canyon on the TE concentrations, including As species, of several common fish species of commercial interest. The hypothesis tested here was that the burden for fish captured close to the Cassidaigne Canyon would be higher for red mud-related trace elements, in particular in Al, Ti and V, than those of fish from other non-impacted areas.

## Materials and methods

### Study area

The Cassidaigne Canyon is a short canyon (<50 km) located in the northern part of the western Mediterranean Sea, east of the Gulf of Lions (Fig. 1). Its head borders Cassis Bay, only 8 km from the coast, and presents in its deeper part two narrow passages located 3 km and 17 km, respectively, from the shelf break (170 m depth). This canyon has been considerably affected by the massive disposal of bauxite residues (Dauvin, 2010) that lasted almost 50 years and is estimated at 20 million tons (ANSES, 2016). Below 350 m, the entire seabed along the canyon axis was covered by the red mud flow which spread more than 60 km from the pipe, with a significant proportion of the mud being propagated at depths >2000 m (Dauvin, 2010). Today, this red mud also covers steep inclined rock and is found underneath overhangs (Fabri et al., 2014). During strong north-eastern Mistral wind events, especially during winter, quantities of this red mud could be transported northward, from the canyon to the continental shelf (Dauvin, 2010).

To assess the impacts of red mud disposal on fish contamination, two zones were considered (Fig. 1). The first was adjacent to the Cassidaigne canyon head (called ‘impacted zone’, IZ), where detectable impact was most likely to occur (see Dauvin, 2010; Fontanier et al., 2012). The second zone (called

76 'reference zone', RZ) was in the vicinity of the Stoechades Canyon. As water masses in this part of the
77 Mediterranean flow westward along the continental margin (Gatti et al., 2006), the Stoechades
78 Canyon is far from the influence of industrial discharge and thus not impacted by bauxite residues
79 (MC. Fabri, pers. com.). The head of the Stoechades Canyon is located in the Gulf of Hyères, less than
80 5 km from the coast, providing similar bathymetry and physico-chemical conditions that allow
81 comparing the two canyons. (Fig. 1).

82

83 Fish sampling

84 Twenty-six commercial species were selected in this study with regard to their importance in the
85 communities of both systems and in the diet of the local population. In addition, these species
86 encompass a wide range of feeding habits and living environments. The sampling can thus be
87 considered representative of all fish functional groups occurring in the area. The aim of the sampling
88 program was to obtain similar species from both the impacted and the reference zone. Fish were
89 collected during 40 fishing operations (16 in the impacted zone, 24 in the reference zone) conducted
90 between June 15$^{th}$ 2015 and July 27$^{th}$ 2015 by professional fishermen using different fishing gears
91 such as bottom and pelagic trawls, trammel nets, hooks, etc. The geographical coordinates of the
92 fishing operations were systematically collected by an on-board scientific observer.

93 Individual fish were stored on board and transferred to the laboratory in ice boxes where all fish
94 were measured (total length to the nearest mm) and weighed (total mass to the nearest 0.1 g)
95 before dissection. Specific measurements were performed in the muscle, the main edible tissue in
96 fish). White dorsal muscle was sampled, individually placed in polyethylene bags and kept frozen at -
97 20°C until chemical analysis. For larger species, namely C*onger conger, Merluccius merluccius,*
98 *Scyliorhinus canicula, Galeus melastomus* and *Raja clavata*, livers were also collected when possible,
99 as liver is the organ mostly involved in detoxication processes.

100

101 Determination of trace element concentrations

102 For each sample, concentrations in 11 TE (Al, As, Cd, Co, Cr, Hg, Mn, Ni, Pb, Ti and V) were analyzed.
103 The analyses of total TE were carried out using a 7700x ICP-MS (Agilent Technologies, Courtaboeuf,
104 France), equipped with a third-generation octopole reaction system (ORS3) using helium as the
105 collision gas. Further details of the instrument settings and performance criteria of this method,
106 validated in-house and accredited by the French Committee of Accreditation (COFRAC), were
107 described in Chevallier et al., (2015). Briefly, 0.2 to 2 g of sample was weighed precisely in a quartz
108 digestion vessel and then wet-oxidised with a mixture of 3 mL of ultra-pure water and 3 mL of ultra-
109 pure $HNO_3$ (67% v/v) in a Multiwave 3000 closed microwave digestion system (Anton-Paar,
110 Courtaboeuf, France). After cooling to room temperature, the digested samples were transferred
111 into 50 mL polyethylene tubes into which a solution of a mixture of internal standards (yttrium,
112 scandium and rhenium) at 2 µg $L^{-1}$ and ultrapure water was added to the final volume before analysis
113 using ICP-MS. One randomly selected vessel was filled with reagents only and taken through the
114 entire procedure as a blank. The limits of detection (LOD) were estimated at 0.042, 0.001, 0.0003,
115 0.0005, 0.005, 0.004, 0.0025, 0.025, 0.0012, 0.025, 0.0005 mg.$kg^{-1}$ wet mass (mg.$kg^{-1}$ wm) whereas
116 the limits of quantification were estimated at 0.083, 0.002, 0.0005, 0.001, 0.010, 0.008, 0.005, 0.050,
117 0.0025, 0.050, 0.0001 mg.$kg^{-1}$ wm for Al, As, Cd, Cr, Hg, Mn, Ni, Pb, Ti and V respectively.

118 Chemical speciation measurements were carried out on the seafood species in which total arsenic
119 exposure was most prevalent: sea breams (gilthead sea bream *Sparus aurata*, black spot sea bream
120 P*agellus bogaraveo*, common two-banded sea bream *Diplodus vulgaris*, white sea bream *Diplodus*
121 *sargus* and red sea bream *Pagellus erythrinus*), redfish and rays. These species contribute more than
122 65% of total arsenic exposure for human consumers. For each species, between 2 and 8 composite
123 samples per zone were analyzed. Each composite sample consisted of 3 to 5 sub-samples. Arsenic
124 speciation was performed by HPLC/ICP-MS (HPLC Ultimate 300, Dionex, Voisins le bretonneux,
125 France; ICP-MS X-Series$^{II}$, Thermo Scientifc, Courtaboeuf, France) coupling in standard mode after

126 microwave-assisted extraction (Multiwave 3000, Anton-Paar). This method was validated in-house,
127 accredited by COFRAC, and described by Leufroy et al. (2011). The following arsenic species were
128 analyzed: As(III), As(V), monomethylarsonic acid (MA), dimethylarsinic acid (DMA), and
129 arsenobetaine (AsB). For all these species, an LOD/LOQ of 0.002/0.005 mg As $kg^{-1}$ wm was assessed
130 under robust conditions. These speciation analyses were used to determine the levels of inorganic
131 arsenic (sum of As(III) and As(V)). In parallel, the total arsenic concentration in the composite
132 samples was also measured by the ICP-MS method described above for total TE analysis.

133 Several internal quality controls (IQCs) and associated criteria were used to ensure the reliability of
134 the results. Each run included a standard calibration, blanks, several certified reference materials
135 (CRMs): TORT-2 lobster hepatopancreas, ERM-CE278k mussel tissue and SRM 1548a typical diet,
136 chosen to cover the 11 elements of interest (for total TE analysis) and BCR-627 tuna fish tissue (for As
137 speciation analysis), two different spiked sample solutions, several fish samples including two
138 samples analyzed in duplicate and a mid-range standard analyzed for every eight samples and at the
139 end of the sequence. The set criteria were as follows: calibration ($r^2 > 0.995$), blanks (values < limit of
140 quantification (LOQ)), internal standards (values within 70 and 130% of the target value), mid-range
141 standards (values within 80 and 120% of the target value), spiked standard solutions (spike recovery
142 within 70 and 130% of the theoretical spiked standard value), CRMs (Z-score < ± 2) and duplicates
143 (acceptable if the relative standard deviation (RSD) ≤ 20% when the mean value ≥ 5 x LOQ or RSD ≤
144 40% when the mean value ≥ LOQ and < 5 x LOQ). When the acceptance criteria were not met, the
145 results were discarded and the samples were re-analyzed (Millour et al., 2010).

146 All concentrations are given in $mg.kg^{-1}$ wm. When data were available in both muscle and liver, the
147 liver to muscle concentration ratios were calculated.

148

149 Statistical analyses

The statistical analysis of the results obtained on both matrices (muscle and liver) of fish was performed only for species presenting a minimum sample size of 15 individuals per zone (IZ and RZ). For these species, Pearson correlation tests were used to investigate the relationship between the TE levels and fish length. Moreover, spatial variations in TE concentrations were analyzed according to the zone for each matrix separately. To do this, PERMANOVAs were performed. They allow handling complex, unbalanced multiple-factor designs and do not assume normal error distributions (Anderson, 2001). Euclidean distance similarity matrices were generated and the factor 'sampling area' was treated as fixed. Since it can affect contamination levels, fish length was considered as a covariate. Data were log(X+1) transformed prior to statistical analyses. In each case, p-values were calculated by 999 random permutations of residuals (Anderson, 2001). Concentrations below LOD were set to 0 whereas concentrations below LOQ were set to LOD. Due to the large number of species and TE analyzed, a large number of statistical analyses were performed simultaneously in this study, thereby increasing the proportion of false positives. Therefore, a multiple testing correction procedure was conducted to control the risk of reporting too many false positives and we used a false discovery rate (FDR) method to adjust for the inflation of p-values (Benjamini and Hochberg, 1995). All data processing and statistical analyses were performed using R software (R Core Team, 2014) and PRIMER 6 software with the PERMANOVA add-on (Clarke and Warwick, 2001). The significance level for the tests was consistently set at $\alpha = 0.05$.

## Results

A total of 1 308 fish of 26 species were collected during this study (769 individuals and 23 species at IZ; 539 individuals and 21 species at RZ, with 18 species collected in both zones) (Table 1, 2).

Considering the two matrices (muscle and liver), 1 597 analyses of 11 trace elements were performed (928 for IZ and 669 for RZ). Among the 26 species sampled, only 9 (*Conger conger, Coris julis, Diplodus vulgaris, Helicolenus dactylopterus, Merluccius merluccius, Mullus surmuletus, Pagellus*

*bogaraveo, Scorpaena porcus* and *Scyliorhinus canicula*) reached the minimum sampling size required to investigate spatial variations and the relationship between TE and fish length. These species represented more than 67% of the total chemical analyses performed. With regards to As speciation, only 66 analyses were performed due to financial constraints (Table 3). This number was too small to correctly explore differences between the two sampling areas and test the effect of fish size on contamination but these results nonetheless provide preliminary information on the burdens of this little investigated TE.

The trace element concentrations in the muscle and the liver of different fish species collected from the two sampling areas are given in Table 2. In muscle, the distribution patterns in the TE concentrations were as follows: As > Al > Hg > Mn > Ti > Pb > Cr > V > Ni > Cd > Co for IZ and As > Al > Hg > Mn > Ti > Pb > Cr > V > Ni > Co > Cd for RZ. In the liver, the sequence was: As > Al > Mn > Hg > Cd > Ti > Pb > Co > V > Cr > Ni for IZ and As > Al > Mn > Hg > Cd > V > Pb > Co > Ti > Cr > Ni for RZ.

Liver to muscle concentration ratios were consistently higher than 1 for Al, Cd, Co, Cr, Mn, Pb and V and reached up to 360 for Cd in *M. merluccius* from RZ. Ratios were lower than 1 (*i.e.* higher concentrations in muscle than in liver) for As and Hg. Due to the high number of values under LOD, it was generally not possible to calculate those ratios for Ni. Finally, the liver to muscle concentration ratios for Ti were below 1 for *C. conger* and *S. canicula* but over 1 for *M. merluccius* and *Raja clavata*.

Contamination levels in muscle differed significantly between species for all TE except Ni (Table 1), and no general specific trend could be observed. Nevertheless, the highest concentrations of As were found in the shark species *S. canicula* ($p < 0.001$) and the highest concentrations of Hg were found in *H. dactylopterus* and *S. canicula* ($p < 0.001$). Considering livers, species-dependent patterns in TE concentrations were less blurred and quite similar between sampling zones, with *S. canicula* presenting the highest Al, As, Cd, Co and Hg concentrations, *M. merluccius* the highest Mn concentrations and *C. conger* the highest Cr, Pb and V concentrations (Table 2).

199 The only compound of inorganic As (Asi) found during this study was AS(III) while As(V)
200 concentrations were systematically below LOD. Inorganic arsenic concentrations in fish muscle were
201 not correlated with total As (AsT) concentrations (Table 3), and the average percentage of Asi
202 (relative to AsT) ranged between 0.01 and 2.69% for all the species considered and in both sampling
203 zones. This percentage of Asi was systematically higher in individuals from IZ than those from RZ.

204 The relationships between fish length and concentrations differed between trace elements, species
205 and tissues (Table 4). The vast majority of relationships, whether significant or not, were nonetheless
206 negative, even for high concentrations of TE in the disposal. On the contrary, Hg was the only TE with
207 a positive relationship for all species and tissues. Finally, As exhibited an intermediate pattern as
208 positive relationships were observed for 8 species × tissue cases studied, 5 of which were significant
209 (muscle of *C. conger, S. canicula* and *P. bogaraveo*, liver of *S. canicula* and *C. conger).*

210 Regarding the spatial trends, TE concentrations varied according to the sampling zone but the nature
211 of the variation differed according to the species and the tissue considered (Table1, 2). Indeed, 36
212 out of the 132 comparisons of TE concentrations between sampling areas remained significant after
213 adjusting for FDR (muscle and liver grouped), with 23 and 13 comparisons displaying significantly
214 higher concentrations in IZ and RZ, respectively. For the main TE from the Gardanne alumina refinery
215 discharges, namely Al, V and Ti, when spatial variations existed, the highest concentrations in fish
216 muscle were systematically observed in IZ. By contrast, for As, Co, Hg, Mn and Pb, no clear pattern
217 was shown with concentrations higher in IZ for some species and higher in RZ for others. Finally, for
218 Cd, Cr and Ni, no difference in TE concentrations were observed between the two zones whatever
219 couple of species × tissue was considered.

220

221 Discussion

222 Very few studies have assessed the impact of the red mud disposal in the Cassidaigne Canyon on
223 contamination levels in fish. To our knowledge, this is the first time that this has been investigated so

224 thoroughly, with the analysis of 11 TE for a large number of individuals (n=1308) from 26 different
225 fish species. In addition, two tissues (muscle and liver) were taken into account, comparing, when
226 possible, contamination levels between the impacted zone and a reference one. Moreover, although
227 several studies have already been conducted on TE in Mediterranean fish (e.g. Canli and Atli, 2003;
228 Iamiceli et al., 2015; Mille et al., 2018), some elements such as Al, Ti and V have rarely been
229 investigated (Eisler, 2010). Lastly, the present study provides a baseline dataset for various
230 Mediterranean fish species and allows a preliminary investigation of certain factors affecting
231 contamination.

232 A large number of the fish analyzed in this study presented concentration levels higher than the
233 limits defined by the European Commission in fish muscle (EC, 2006). A total of 444 individuals from
234 15 different species (*C. conger, C. julis, D. sargus, D. vulgaris, G. melastomus, H. dactylopterus,*
235 *M. merluccius, M. surmuletus, M. barbatus, P. bogaraveo, R. clavata, S. canicula, T. mediterraneus,*
236 *T. trachurus and Z. faber)* had Hg concentration kevels above the European regulatory limit (0.5
237 $mg.kg^{-1}$ wm in teleosts, 1 $mg.kg^{-1}$ wm in sharks), confirming the concern raised previously regarding
238 Hg in Mediterranean organisms (Cossa et al., 2012; Harmelin-Vivien et al., 2009; Koenig et al., 2013).
239 The straightforward pattern observed for Hg in the present study may be largely driven by biological
240 and biogeochemical mechanisms at play at the scale of the whole NE Mediterranean (Cossa and
241 Coquery, 2005) rather than from the local influence of red mud disposals. For Pb and Cd, the number
242 of individuals with concentrations above the EU regulatory limits in fish muscle (0.3 $mg.kg^{-1}$ wm and
243 0.05 $mg.kg^{-1}$ wm for Pb and Cd, respectively), were lower, 8 and 19, respectively, which represent
244 between 0.6 and 1.5% of the total number of fish analyzed. Between 75 and 90% of these threshold
245 overruns concerned IZ, confirming the conclusions of Ourgaud et al. (2018), who found that these TE
246 were more concentrated in *S. porcus* collected in the bay of Marseille (close to the Cassidaigne
247 Canyon) than in those from the Gulf of Hyères. There is no regulatory limit for total As in fish muscle,
248 but arsenic exposure is a significant worldwide environmental health concern. Overall, the data we
249 obtained here appear of great concern with regard to total As concentrations. However, the risk of

potential As poisoning through fish consumption should be further assessed through the specific determination of the inorganic As content in fish tissues. Most of the As in the tissues of marine animals is bound in organoarsenic forms (Neff, 1997) and considered of low toxicity (Hindmarsh, 2000). In this study, the main form of As found in fish was arsenobetaine which accounts for up to 100% of As in certain species (Table 3). This form is not toxic to humans (Neff, 1997).

In both sampling zones, our results clearly showed significant differences in the bioaccumulation of a range of TE between the 26 selected species, suggesting a diversity response among fish (Table 1, 2). Indeed, the accumulation of TE in fish does not directly reflect their simple concentration in the ambient environment as it is affected by environmental factors, species biology and the physico-chemical properties of the contaminant (Cossa et al., 2012; Cresson et al., 2015). Thus, assessing a site-specific contamination pattern requires good understanding of all these parameters although the interpretation of the results remains complex because of the existence of interactions between them. Notwithstanding some general trends could still be discerned. Some TE such as Hg are well-known for their biomagnification property, namely the trend for increasing concentrations along food webs (Bryan et al., 1979). This process in particular is responsible for the high Hg concentrations generally observed in high trophic level species such as marine apex predators (Endo et al., 2008) like *S. canicula*, *M. merluccius*, *G melastomus*, *Z. faber*, etc. In this study, Hg concentrations in fish muscle from IZ varied between 0.014 $mg.kg^{-1}$ in *L. aurata* and 1.613 $m. kg^{-1}$ in *S. canicula* (Table 1). The inter-specific variation in mean trophic levels, ranging from 2.8 for *L. aurata* to 3.8 *S. canicula* (Cresson et al., 2014b; Froese and Pauly, 2014; Stergiou and Karpouzi, 2001), could partially explain this differences in the TE concentrations between these species. Moreover, an increasing number of studies have documented relatively high levels of TE contamination in shelf-edge and deep species (Cresson et al., 2014a; Hornung et al., 1993), especially when compared to coastal species (Chouvelon et al., 2012). The 26 species selected here live at drastically different depths, from the surface down to a few meters depth *for C. julis*, *L. ramada* and *L. aurata,* to several hundreds of meters for *G. melastomus*, *H. dactylopterus* and *P. bogaraveo* (Froese and Pauly, 2014). In this study,

276 *C. julis*, *L. ramada* and *L. aurata* were captured between 5 and 35 m depth whereas *G. melastomus*,
277 *H. dactylopterus* and *P. bogaraveo* were caught between 141 and 391m depth. This may explain a
278 part of the inter-specific variations in TE contamination and the lower TE concentrations generally
279 observed in *L. ramada*, *L. aurata* and *C. julis* (Table 1), Finally, inter-specific differences in TE
280 concentrations may also be driven by contrasting life-history and behavior. Factors such as preferred
281 habitat (*e.g*. benthic and pelagic), longevity, diet and foraging behavior (*e. g.* vertical migrations and
282 type of prey consumed), metabolic rates and biochemical response (Burger, 2007; Zhao et al., 2012)
283 have been demonstrated to affect rates of TE uptake and excretion from both food and water
284 pathways. Previous works showed that species that are continuously in contact with contaminants in
285 sediment have developed higher detoxication abilities, resulting in lower body burdens while being
286 exposed to the same contaminant load as pelagic species (Sole et al., 2006). This may explain why TE
287 concentrations in the benthic ray *R. clavata* are genarally lower than those o the demersal *S. canicula*
288 (Table1, 2), despite similar trophic levels (Froese and Pauly, 2014; Stergiou and Karpouzi, 2001) and
289 living depth (both species were captured between 100 and 380m depth).

290 Fish length is often recognized to be of importance in determining the rate of physiological processes
291 that influence uptake, distribution and elimination of TE (Canli and Atli, 2003). However, except for
292 Hg, our analysis revealed no conclusive evidence that mean TE concentrations were systematically
293 linked to fish length, which is in good agreement with the results reported in the literature. Indeed,
294 although it is generally accepted that Hg load in fish body is strongly affected by size it is still not
295 clear if the other TE accumulations are size-dependent. This is supported by the fact that most TE
296 (with the exception of As and Hg) do not biomagnify along aquatic food webs (Fey et al., 2019; Xu
297 and Wang, 2002). The available literature on the correlation between TE concentrations in muscle
298 tissue and fish length and weight have shown that a dependency exists for some species (e.g. Gobert
299 et al., 2017), while for other species, no dependency is found (e.g. Mille et al., 2018). The absence of
300 relationship likely indicates that fish regulate some TE at a certain concentration required for
301 efficient metabolic activities (Canli and Atli, 2003; Hornung et al., 1993). Moreover, when significant,

302 the relationships between TE (other than Hg) and fish lengths are generally negative (Endo et al.,
303 2008). This may result from a higher metabolic activity in fast-growing young individuals, requiring
304 higher amounts of these elements than older individuals or through a higher excretion rate and/or
305 dilution of metal burden with growth, and lower metabolism (Eisler, 2010). In addition, length is
306 commonly used as a proxy of age and chronic exposure to contaminants, as length is quickly
307 measured, while accurate age determination requires complex and time-consuming procedures (*e.g.*
308 otolith or vertebrae reading for teleost and chondrichtyans, respectively). This pattern is nonetheless
309 blurred by the fact that growth rate affects the age - length relationship. As a result, individuals of
310 the same length but from zones with different nutritional inputs (and thus different growth rates)
311 may have different age and may have been thus exposed to contaminant during different periods of
312 time.

313 Many studies have shown that TE accumulate in various organs of fish at different levels (Canli and
314 Atli, 2003; Dural et al., 2006; Fernandes et al., 2007; Long and Wang, 2005; Mormede and Davies,
315 2001; Uluturhan and Kucuksezgin, 2007). With the exception of Hg, As and Ti, the liver to muscle
316 concentration ratios calculated in this study were generally over 1 and reached up to 360. This is not
317 surprising because, in general, TE are preferentially accumulated in the liver of aquatic organisms
318 (Henry et al., 2004), which likely results from detoxification processes (Eisler, 2010). For Hg, higher
319 concentrations are commonly found in fish muscle. Indeed, methylmercury is the predominant form
320 of Hg in fish, ranging from 80% to 100% (Bloom, 1992; Chouvelon et al., 2018; Magalhaes et al.,
321 2007) and is mainly accumulated in fish muscle due to its affinity to protein (Amlund et al., 2007;
322 Harris et al., 2003). This difference in chemical affinity explains why the liver to muscle concentration
323 ratios for Hg ranged here from 0.47 to 0.97. Considering As, Eisler (2010) stated that hepatic
324 concentrations are usually higher than those found in muscle tissues, a pattern not observed in the
325 present study as the liver to muscle concentration ratios ranged between 0.34 and 0.90. Such ratios
326 below 1 have already been reported in benthic (*Pteromylaeus bovinus* and *Myliobatis aquila*) and
327 pelagic (*Pteroplatytrygon violacea*) rays from the northern Adriatic Sea (Slejkovec et al., 2014), in *S.*

328 *canicula* from the North Sea (De Gieter et al., 2002), in 22 fish species from the New Caledonia
329 lagoon (Metian et al., 2013), and in 32 teleost species from the island of Guam (Pacific Ocean)
330 (Denton et al., 2006). Therefore, the As partition between muscle and liver seems to be species-
331 dependent (e.g. Hellou et al., 1996; Raimundo et al., 2013; Raimundo et al., 2015) and possibly
332 related to differences in the bioavailability of forms of this element (Francesconi, 2010). Finally, very
333 few data exist on Ti concentrations in fish and, to our knowledge, the accumulation of this element in
334 different organs has never been studied. The distinct accumulation patterns observed here may
335 result from differences in the physiological response of the species, as well as differences in life-
336 history and behavior of fish.

337 As confirmed by sediment analysis (Fontanier et al., 2012), the Cassidaigne Canyon has been
338 subjected to considerable TE inputs, mainly in Al, Ti, V from the alumina refinery's by-products. In
339 general, these TE are considered non-essential for marine life and their concentrations in organisms
340 mainly depend on their environmental levels (Amiard et al., 1987). Therefore, it was expected that
341 fish from IZ would contain consistently higher concentrations of red mud-related TE than those from
342 RZ. Among all the species, Al was consistently more accumulated in fish from IZ, likely resulting from
343 industrial releases. Aluminum is also the TE most concentrated in red mud. Laboratory experiments
344 showed that the bauxite from the Gardanne plant releases substantial quantities of Al in seawater
345 (Pagano et al., 2002). In the marine environment (pH 8.0-8.3, salinity 35), this Al is theoretically
346 mostly present in two forms: the aluminate anion $Al(OH)_4^-$ and to a lesser extent neutral aluminum
347 hydroxide $Al(OH)_3$ (Millero et al., 2009). There are very few studies on the transfer of aluminate
348 anions and aluminum hydroxide to aquatic organisms and even fewer focusing on fish species. Our
349 results suggested that fish are able to at least partially accumulate these Al forms. This observation
350 can be extended to other living organisms as Al bioaccumulation has also been reported in marine
351 plants collected nearby a coastal bauxite sludge disposal site (Malea and Haritonidis, 1989). For the
352 other TE attributable to the Gardanne alumina refinery discharges (Ti and V), we found that when
353 spatial variations existed, concentrations in fish muscle were systematically higher in IZ than in RZ,

but this difference concerned a limited number of species. It could be hypothesized that they are not readily bioavailable for species and/or they are poorly transferred through the food chain up to apex predators. This result is similar to those of Bourcier (1969), Bourcier & Zibrowius (1973) and Fontanier et al. (2012), who observed that there was no chemical impact of red muds on macrobenthic fauna and the structure of deep-sea foraminifera communities in the Cassidaigne Canyon. They suggested that most of the toxic TE in the red mud impacting the sites are not bioavailable but simply locked in solid phases.

## Conclusion

The impact of the human activities on the levels of fish contamination by TE is not easy to assess *in situ* because it is blurred by the interaction of biological effects (e.g. Bouchoucha et al., 2018; Cresson et al., 2014a; Metian et al., 2013). A thorough sampling campaign and numerous analyses allowed highlighting the significant contamination of fish species collected from the Cassidaigne Canyon area, especially for Al and to a lesser extent for Ti and V. Although not statistically proved, it was also shown that fish collected close to the alumina refinery pipe had higher Asi concentrations than those from the reference area. In the next step, it would be interesting to assess human dietary exposure to these metals through the consumption of fish from the Cassidaigne Canyon zone. This would allow better evaluation of the health risk for humans consuming these fish. Moreover, the solubility of many TE is highly redox-sensitive, thus the elemental concentrations in seawater change accordingly (e.g. Morford and Emerson, 1999). Consequently, if the redox conditions change in the sites contaminated by red mud, this would influence the solubility of TE, making them bioavailable to marine organisms. Therefore, it seems essential to continue regular and long-term monitoring of marine organisms from this area to prevent any further environmental deterioration and to assess human exposure.

Acknowledgements

This work received financial support from the Environment and Sustainable Development Ministry.

We are grateful to F. Ruchon and local fishermen for their help in fish sampling. Thanks are also due to C. Tomasino, D. Louedoc, C. Ravel, A. Vaugier and F. Chavanon for their assistance during the laboratory work.

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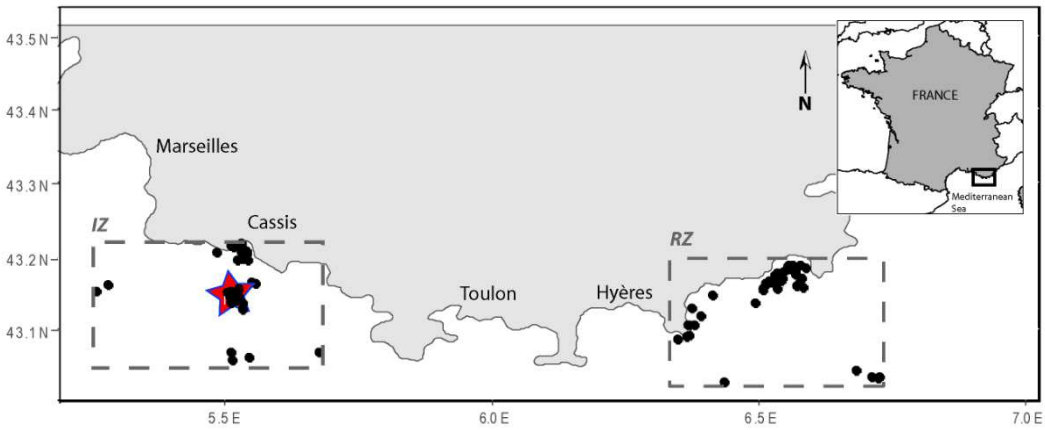
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Figures

**Figure 1:** Location of the sampling station and sampling zones. The star represents the Gardanne alumina refinery pipeline outlet.



658 **Table 1:** Mean TE concentrations ± standard deviation (in $mg.kg^{-1}$ wm) measured in fish muscle from the red mud impacted (IZ) and the reference (RZ)
659 zones. Minimum and maximum values are given in brackets.
660

| Species | zone | Tissue | N | Total length (mm) | Total mass (g) | Al | As | Cd | Co | Cr | Hg |
|---|---|---|---|---|---|---|---|---|---|---|---|
| *Chelon labrosus* | IZ | muscle | 1 | 406 | 860.3 | 0.320 | 1.659 | 0.000 | 0.002 | 0.021 | 0.128 |
| *Conger conger* | IZ | muscle | 31 | 710 ± 171 (416 - 1119) | 799.5 ± 643.2 (94.5 – 2894.4) | 0.062 ± 0.159 (0.000 - 0.858) | 22.115 ± 11.886 (5.308 - 53.193) | 0.003 ± 0.007 (0.000 - 0.038) | 0.002 ± 0.003 (0.000 - 0.015) | 0.021 ± 0.029 (0.000 - 0.109) | 0.38 ± 0.339 (0.07 - 1.872) |
| *Conger conger* | RZ | muscle | 23 | 716 ± 203 (334 - 1035) | 817.5 ± 672.6 (50.8- 2341.7) | 0.032 ± 0.102 (0.000 - 0.465) | 26.867 ± 19.009 (8.613 - 86.156) | 0.003 ± 0.008 (0.000 - 0.041) | 0.001 ± 0.001 (0.000 - 0.003) | 0.017 ± 0.023 (0.000 - 0.085) | 0.345 ± 0.218 (0.066 - 1.003) |
| *Coris julis* | IZ | muscle | 32 | 128 ± 1.9 (103 - 212) | 20.9 ± 12.5 (8.6 – 83.5) | 0.106 ± 0.112 (0.000 - 0.389) | 4.213 ± 2.266 (1.08 - 10.100) | 0.001 ± 0.001 (0.000 - 0.002) | 0.007 ± 0.003 (0.000 - 0.017) | 0.031 ± 0.035 (0.000 - 0.145) | 0.148 ± 0.105 (0.095 - 0.700) |
| *Coris julis* | RZ | muscle | 31 | 125 ± 14 (86 - 147) | 17.0 ± 5.3 (5.1 - 29.4) | 0.149 ± 0.237 (0.000 - 1.11) | 7.633 ± 7.786 (0.586 - 33.938) | 0.004 ± 0.015 (0.000 - 0.085) | 0.009 ± 0.004 (0.000 - 0.019) | 0.014 ± 0.019 (0.000 - 0.065) | 0.174 ± 0.245 (0.035 - 1.369) |
| *Dicentrarchus labrax* | RZ | muscle | 1 | 349 | 346.6 | 0.000 | 0.675 | 0.000 | 0.002 | 0.027 | 0.222 |
| *Diplodus sargus* | IZ | muscle | 4 | 269 ± 21 (235 - 289) | 357.2 ± 86.9 (214.6 - 433.3) | 0.026 ± 0.051 (0.000 - 0.102) | 5.28 ± 2.908 (2.203 - 9.221) | 0.001 ± 0.000 (0.001 - 0.001) | 0.004 ± 0.001 (0.003 - 0.004) | 0.016 ± 0.012 (0.000 - 0.028) | 0.756 ± 0.364 (0.429 - 1.264) |
| *Diplodus sargus* | RZ | muscle | 39 | 245 ± 45 (173 - 375) | 308.7 ± 189.1 (101 - 969.4) | 0.385 ± 1.784 (0.000 - 11.076) | 11.911 ± 6.165 (1.218 - 24.651) | 0.001 ± 0.001 (0.000 - 0.005) | 0.007 ± 0.008 (0.000 - 0.047) | 0.009 ± 0.016 (0.000 - 0.073) | 0.429 ± 0.296 (0.084 - 1.369) |
| *Diplodus vulgaris* | IZ | muscle | 28 | 226 ± 20 (189-279) | 203.5 ± 59.0 (83.6 - 359.4) | 0.262 ± 0.154 (0.000 - 0.683) | 4.346 ± 1.188 (1.585 - 7.514) | 0.001 ± 0.005 (0.000 - 0.024) | 0.003 ± 0.002 (0.000 - 0.010) | 0.016 ± 0.045 (0.000 - 0.228) | 0.442 ± 0.176 (0.124 - 0.786) |
| *Diplodus vulgaris* | RZ | muscle | 53 | 213 ± 24 (149 - 249) | 164.0 ± 56.8 (46.9 - 283.9) | 0.052 ± 0.083 (0.000 - 0.304) | 6.919 ± 3.132 (3.209 - 15.573) | 0.001 ± 0.001 (0.000 - 0.003) | 0.005 ± 0.002 (0.000 - 0.011) | 0.016 ± 0.019 (0.000 - 0.096) | 0.389 ± 0.159 (0.082 - 0.798) |
| *Engraulis encrasicolus* | IZ | muscle | 60 | 139 ± 8 (121-157) | 16.8 ± 2.9 (10.2 - 25.5) | 0.030 ± 0.074 (0.000 - 0.363) | 3.804 ± 0.759 (1.688 - 5.900) | 0.005 ± 0.002 (0.002 - 0.009) | 0.01 ± 0.002 (0.006 - 0.017) | 0.019 ± 0.018 (0.000 - 0.059) | 0.119 ± 0.046 (0.035 - 0.247) |
| *Engraulis encrasicolus* | RZ | muscle | 6 | 118 ± 11 (100 - 129) | 10.6 ± 2.7 (5.5 - 12.5) | 1.769 ± 2.503 (0.160 - 5.711) | 4.664 ± 0.846 (3.830 - 6.252) | 0.005 ± 0.002 (0.000 - 0.007) | 0.016 ± 0.004 (0.011 - 0.022) | 0.035 ± 0.019 (0.017 - 0.070) | 0.084 ± 0.02 (0.065 - 0.118) |
| *Galeus melastomus* | IZ | muscle | 32 | 459 ± 81 (323 - 578) | 283.4 ± 147.3 (81.8 - 545.7) | 0.574 ± 0.583 (0.000 - 3.090) | 33.122 ± 7.735 (22.517 - 55.173) | 0.001 ± 0.002 (0.000 - 0.006) | 0.001 ± 0.002 (0.000 - 0.007) | 0.019 ± 0.017 (0.000 - 0.074) | 1.47 ± 0.842 (0.553 - 3.833) |
| *Galeus melastomus* | RZ | muscle | 4 | 327 ± 31 (277 - 354) | 89.8 ± 23.1 (51.2 - 110.0) | 1.923 ± 1.016 (0.812 - 2.829) | 28.503 ± 9.637 (17.587 - 41.050) | 0.001 ± 0.000 (0.001 - 0.001) | 0.003 ± 0.001 (0.002 - 0.005) | 0.038 ± 0.005 (0.031 - 0.042) | 0.908 ± 0.139 (0.799 - 1.089) |
| *Helicolenus dactylopterus* | IZ | muscle | 37 | 187 ± 31 (114 - 248) | 109.6 ± 51.2 (22.2 - 264.9) | 0.694 ± 0.683 (0.137-3.364) | 5.281 ± 2.442 (2.111 - 12.100) | 0.001 ± 0.001 (0.000 - 0.006) | 0.001 ± 0.002 (0.000 - 0.005) | 0.029 ± 0.086 (0.000 - 0.525) | 1.045 ± 0.456 (0.273 - 2.730) |
| *Helicolenus dactylopterus* | RZ | muscle | 46 | 194 ± 33 (139-291) | 130.7 ± 72.2 (37.6 - 402) | 0.217 ± 0.301 (0.000 - 1.839) | 10.166 ± 5.484 (2.690 - 25.008) | 0.001 ± 0.001 (0.000 - 0.003) | 0.003 ± 0.002 (0.000 - 0.010) | 0.013 ± 0.024 (0.000 - 0.137) | 1.099 ± 0.368 (0.382 - 2.059) |
| *Liza aurata* | IZ | muscle | 12 | 281 ± 24 (241-330) | 193.3 ± 71.1 (111.7 - 388.8) | 0.064 ± 0.084 (0.000 - 0.217) | 0.561 ± 0.253 (0.285 - 0.960) | 0.000 ± 0.000 (0.000 - 0.000) | 0.01 ± 0.002 (0.006 - 0.013) | 0.027 ± 0.025 (0.000 - 0.076) | 0.014 ± 0.01 (0.000 - 0.028) |
| *Liza aurata* | RZ | muscle | 7 | 328 ± 25 (282 - 359) | 307.3 ± 65.6 (212.6 - 385.9) | 0.050 ± 0.089 (0.000 - 0.218) | 1.302 ± 0.477 (0.832 - 2.230) | 0.000 ± 0.000 (0.000 - 0.000) | 0.011 ± 0.006 (0.000 - 0.018) | 0.024 ± 0.036 (0.000 - 0.088) | 0.021 ± 0.012 (0.013 - 0.047) |
| *Liza ramada* | IZ | muscle | 13 | 319 ± 50 (239 - 389) | 288.7 ± 144.8 (105.7 - 506.2) | 0.591 ± 0.447 (0.101 - 1.770) | 0.992 ± 0.589 (0.243 - 2.091) | 0.000 ± 0.000 (0.000 - 0.000) | 0.006 ± 0.003 (0.002 - 0.010) | 0.019 ± 0.019 (0.000 - 0.06) | 0.094 ± 0.095 (0.007 - 0.321) |
| *Merluccius merluccius* | IZ | muscle | 53 | 354 ± 132 (233 - 795) | 439.1 ± 518.4 (89.1 - 2305.2) | 0.460 ± 0.474 (0.000 - 2.139) | 6.512 ± 2.707 (3.353 - 18.323) | 0.002 ± 0.007 (0.000 - 0.043) | 0.001 ± 0.005 (0.000 - 0.021) | 0.005 ± 0.011 (0.000 - 0.041) | 0.454 ± 0.524 (0.08 - 3.073) |

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|---|---|---|---|---|---|---|---|---|---|---|---|
| *Merluccius merluccius* | RZ | muscle | 24 | 384 ± 54 (280 - 476) | 456.9 ±185.6 (147.3 – 858.4) | 0.131 ± 0.16 (0.000 - 0.487) | 5.070 ± 2.194 (2.405 - 11.150) | 0.000 ± 0.001 (0.000 - 0.004) | 0.000 ± 0.000 (0.000 - 0.002) | 0.006 ± 0.009 (0.000 - 0.026) | 0.497 ± 0.39 (0.104 - 1.593) |
| *Mugil cephalus* | IZ | muscle | 1 | 425 | 969.5 | 0.105 | 3.597 | 0.000 | 0.003 | 0.061 | 0.327 |
| *Mullus barbatus* | IZ | muscle | 77 | 171 ± 25 (121 - 230) | 57.7 ± 27.7 (11.7 – 140.7) | 1.209 ± 1.434 (0.000 - 9.152) | 24.775 ± 7.771 (1.43 - 50.586) | 0.001 ± 0.002 (0.000 - 0.013) | 0.008 ± 0.004 (0.003 - 0.022) | 0.017 ± 0.02 (0.000 - 0.091) | 0.46 ± 0.338 (0.06 - 1.743) |
| *Mullus barbatus* | RZ | muscle | 2 | 212 ± 28 (185 - 239) | 115.0 ± 45.6 (70.4 – 159.5) | 0.203 ± 0.163 (0.088 - 0.318) | 13.222 ± 10.906 (5.510 - 20.933) | 0.000 ± 0.000 (0.000 - 0.000) | 0.007 ± 0.002 (0.005 - 0.008) | 0.044 ± 0.037 (0.018 - 0.07) | 0.217 ± 0.247 (0.042 - 0.391) |
| *Mullus surmuletus* | IZ | muscle | 68 | 181 ± 29 (135 - 267) | 75.2 ± 41.1 (27.2 – 236.1) | 1.294 ± 3.004 (0.116 - 24.632) | 15.112 ± 5.732 (5.647 - 34.549) | 0.000 ± 0.001 (0.000 - 0.004) | 0.005 ± 0.002 (0.002 - 0.016) | 0.014 ± 0.015 (0.000 - 0.065) | 0.426 ± 0.51 (0.102 - 2.585) |
| *Mullus surmuletus* | RZ | muscle | 72 | 188 ± 30 (152 - 315) | 88.4 ± 57.9 (39.1 – 418.8) | 0.268 ± 0.453 (0.000 - 2.32) | 11.828 ± 5.166 (3.090 - 28.257) | 0.000 ± 0.000 (0.000 - 0.002) | 0.004 ± 0.003 (0.000 - 0.012) | 0.014 ± 0.022 (0.000 - 0.112) | 0.132 ± 0.098 (0.022 - 0.403) |
| *Pagellus bogaraveo* | IZ | muscle | 27 | 268 ± 19 (236 - 307) | 267.9 ± 63.6 (149.6 – 420.6) | 0.328 ± 0.436 (0.000 - 2.083) | 5.87 ± 2.535 (2.250 - 11.900) | 0.001 ± 0.002 (0.000 - 0.010) | 0.004 ± 0.003 (0.000 - 0.012) | 0.009 ± 0.015 (0.000 - 0.066) | 0.318 ± 0.174 (0.092 - 1.041) |
| *Pagellus bogaraveo* | RZ | muscle | 30 | 240 ± 10 (221 - 265) | 204.5 ± 24.4 (167.5 – 273.8) | 0.203 ± 0.157 (0.000 - 0.671) | 5.417 ± 1.138 (3.212 - 7.746) | 0.001 ± 0.001 (0.000 - 0.002) | 0.006 ± 0.002 (0.003 - 0.014) | 0.017 ± 0.041 (0.000 - 0.220) | 0.329 ± 0.194 (0.183 - 0.995) |
| *Pagellus erythrinus* | RZ | muscle | 6 | 172 ± 11 (154 - 188) | 68.3 ± 10.7 (55.1 – 83.1) | 0.948 ± 0.191 (0.743 - 1.27) | 5.06 ± 1.275 (3.640 - 6.350) | 0.000 ± 0.000 (0.000 - 0.000) | 0.002 ± 0.003 (0.000 - 0.006) | 0.003 ± 0.007 (0.000 - 0.016) | 0.171 ± 0.049 (0.091 - 0.234) |
| *Raja clavata* | IZ | muscle | 21 | 605 ± 116 (440 - 826) | 1493.4 ± 940.0 (431.4 – 3820.1) | 0.559 ± 1.143 (0.000 - 4.987) | 76.051 ± 30.462 (1.404 - 119.085) | 0.099 ± 0.300 (0.001 - 1.325) | 0.013 ± 0.025 (0.000 - 0.088) | 0.005 ± 0.009 (0.000 - 0.029) | 1.185 ± 0.723 (0.156 - 3.765) |
| *Raja clavata* | RZ | muscle | 5 | 749 ± 68 (669 - 849) | 2293.3 ± 650.1 (1567.7 – 3181.4) | 0.080 ± 0.129 (0.000 - 0.296) | 89.171 ± 22.568 (68.400 - 119.268) | 0.001 ± 0.001 (0.001 - 0.002) | 0.002 ± 0.001 (0.000 - 0.003) | 0.015 ± 0.014 (0.000 - 0.028) | 1.429 ± 0.74 (0.739 - 2.323) |
| *Sardina pilchardus* | IZ | muscle | 68 | 141 ± 11 (124 - 193) | 21.3 ± 5.9 (12.5 – 54.7) | 0.393 ± 0.968 (0.000 - 7.817) | 5.805 ± 1.341 (4.028 - 12.264) | 0.001 ± 0.002 (0.000 - 0.007) | 0.011 ± 0.002 (0.007 - 0.016) | 0.009 ± 0.014 (0.000 - 0.073) | 0.1 ± 0.042 (0.046 - 0.334) |
| *Sardinella aurita* | RZ | muscle | 2 | 240 ± 6 (234 - 246) | 101.3 ± 6.9 (94.5 – 108.0) | 0.068 ± 0.096 (0.000 - 0.136) | 6.230 ± 1.061 (5.480 - 6.980) | 0.001 ± 0.000 (0.001 - 0.001) | 0.003 ± 0.004 (0.000 - 0.006) | 0.007 ± 0.009 (0.000 - 0.013) | 0.103 ± 0.000 (0.103 - 0.103) |
| *Scorpaena porcus* | IZ | muscle | 39 | 187 ± 39 (129 - 262) | 155.2 ± 99.7 (45.1 – 392.6) | 0.808 ± 0.861 (0.000-3.689) | 4.190 ± 2.439 (0.735 - 11.700) | 0.001 ± 0.001 (0.000 - 0.006) | 0.001 ± 0.001 (0.000 - 0.005) | 0.013 ± 0.02 (0.000 - 0.096) | 0.151 ± 0.049 (0.072 - 0.307) |
| *Scorpaena porcus* | RZ | muscle | 71 | 160 ± 21 (119 - 218) | 83.6 ± 37.4 (33.7 – 201.7) | 0.778 ± 0.745 (0.108 - 3.74) | 4.826 ± 3.0590 (0.577 - 12.600) | 0.001 ± 0.001 (0.000 - 0.006) | 0.003 ± 0.004 (0.000 - 0.015) | 0.012 ± 0.026 (0.000 - 0.153) | 0.171 ± 0.061 (0.072 - 0.361) |
| *Scyliorhinus canicula* | IZ | muscle | 63 | 417 ± 59 (244 - 509) | 245.1 ± 90.8 (39.4 – 400.4) | 1.259 ± 1.727 (0.196 - 11.497) | 37.735 ± 17.946 (15.030 - 112.802) | 0.065 ± 0.221 (0.000 - 1.541) | 0.012 ± 0.026 (0.000 - 0.163) | 0.032 ± 0.026 (0.000 - 0.144) | 1.613 ± 1.335 (0.233 - 8.955) |
| *Scyliorhinus canicula* | RZ | muscle | 82 | 370 ± 62 (244 - 500) | 183.6 ± 103.9 (41.3 – 485.4) | 1.299 ± 0.995 (0.000 - 4.815) | 41.341 ± 19.253 (0.020 - 118.615) | 0.002 ± 0.003 (0.000 - 0.026) | 0.007 ± 0.005 (0.000 - 0.034) | 0.042 ± 0.034 (0.000 - 0.254) | 0.845 ± 0.309 (0.000 - 1.862) |
| *Sparus aurata* | IZ | muscle | 24 | 252 ± 15 (232 - 306) | 233.9 ± 46.1 (183.6 – 419.6) | 0.031 ± 0.075 (0.000 - 0.254) | 4.816 ± 3.0760 (1.263 - 10.910) | 0.000 ± 0.000 (0.000 - 0.002) | 0.004 ± 0.002 (0.002 - 0.009) | 0.009 ± 0.016 (0.000 - 0.069) | 0.15 ± 0.087 (0.077 - 0.411) |
| *Sparus aurata* | RZ | muscle | 4 | 261 ± 45 (223 - 336) | 264.7 ± 150.2 (150.2 – 519.6) | 0.147 ± 0.033 (0.117 - 0.189) | 11.948 ± 8.455 (4.940 - 23.710) | 0.000 ± 0.000 (0.000 - 0.000) | 0.003 ± 0.001 (0.002 - 0.005) | 0.009 ± 0.011 (0.000 - 0.024) | 0.193 ± 0.166 (0.056 - 0.421) |
| *Trachurus mediterraneus* | IZ | muscle | 8 | 331 ± 20 (307 - 366) | 283.4 ± 49.6 (238 - 393.6) | 0.441 ± 0.420 (0.000 - 1.081) | 1.593 ± 1.122 (0.549 - 3.889) | 0.002 ± 0.004 (0.000 - 0.011) | 0.005 ± 0.001 (0.003 - 0.008) | 0.023 ± 0.024 (0.000 - 0.074) | 0.511 ± 0.133 (0.342 - 0.741) |
| *Trachurus mediterraneus* | RZ | muscle | 30 | 272 ± 22 (247 - 318) | 165.2 ± 41.8 (115.3 – 256.8) | 0.298 ± 0.260 (0.000 - 0.921) | 1.815 ± 1.113 (0.735 - 4.63) | 0.001 ± 0.002 (0.000 - 0.009) | 0.007 ± 0.005 (0.000 - 0.026) | 0.011 ± 0.019 (0.000 - 0.071) | 0.378 ± 0.135 (0.194 - 0.697) |
| *Trachurus trachurus* | IZ | muscle | 34 | 337 ± 37 (281 - 425) | 352.1 ± 130.8 (180 – 659.1) | 0.176 ± 0.110 (0.000 - 0.427) | 5.429 ± 2.0460 (1.478 - 10.51) | 0.002 ± 0.005 (0.000 - 0.029) | 0.008 ± 0.004 (0.000 - 0.022) | 0.015 ± 0.03 (0.000 - 0.173) | 0.947 ± 0.334 (0.399 - 1.88) |
| *Zeus faber* | IZ | muscle | 36 | 322 ± 96 (131 - 511) | 581.6 514.5 (38.1 – 2278.2) | 0.142 ± 0.340 (0.000 - 2.030) | 0.619 ± 0.308 (0.302 - 1.406) | 0.001 ± 0.004 (0.000 - 0.019) | 0.001 ± 0.001 (0.000 - 0.004) | 0.013 ± 0.03 (0.000 - 0.133) | 0.569 ± 0.496 (0.046 - 2.251) |

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|---|---|---|---|---|---|---|---|---|---|---|---|
| *Zeus faber* | RZ | muscle | 1 | 355 | 612.2 | 0.130 | 0.469 | 0.000 | 0.002 | 0.101 | 0.25 |

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663 **Table 1 (continued):** Mean TE concentrations ± standard deviation (in mg.kg$^{-1}$ wm) measured in fish muscle from the zone (IZ) impacted by red mud and the
664 reference (RZ) zones. Minimum and maximum values are given in brackets.
665

| Species | zone | tissue | N | Total length (mm) | Total mass (g) | Mn | Ni | Pb | Ti | V |
|---|---|---|---|---|---|---|---|---|---|---|
| *Chelon labrosus* | IZ | muscle | 1 | 406 | 860.3 | 0.069 | 0.000 | 0.005 | 0.001 | 0.001 |
| *Conger conger* | IZ | muscle | 31 | 710 ± 171 (416 - 1119) | 799.5 ± 643.2 (94.5 – 2894.4) | 0.346 ± 0.260 (0.07 - 1.035) | 0.000 ± 0.000 (0.000 - 0.000) | 0.007 ± 0.007 (0.000 - 0.031) | 0.067 ± 0.114 (0.000 - 0.54) | 0.006 ± 0.0140 (0.000 - 0.077) |
| *Conger conger* | RZ | muscle | 23 | 716 ± 203 (334 - 1035) | 817.5 ± 672.6 (50.8- 2341.7) | 0.515 ± 0.383 (0.058 - 1.273) | 0.000 ± 0.000 (0.000 - 0.000) | 0.006 ± 0.008 (0.000 - 0.031) | 0.070 ± 0.115 (0.000 - 0.466) | 0.004 ± 0.004 (0.000 - 0.017) |
| *Coris julis* | IZ | muscle | 32 | 128 ± 1.9 (103 - 212) | 20.9 ± 12.5 (8.6 – 83.5) | 0.182 ± 0.058 (0.097 - 0.304) | 0.007 ± 0.024 (0.000 - 0.109) | 0.025 ± 0.019 (0.000 - 0.076) | 0.059 ± 0.06 (0.000 - 0.254) | 0.030 ± 0.017 (0.000 - 0.090) |
| *Coris julis* | RZ | muscle | 31 | 125 ± 14 (86 - 147) | 17.0 ± 5.3 (5.1 - 29.4) | 0.196 ± 0.074 (0.107 - 0.415) | 0.003 ± 0.011 (0.000 - 0.051) | 0.014 ± 0.012 (0.000 - 0.043) | 0.114 ± 0.127 (0.000 - 0.513) | 0.048 ± 0.043 (0.007 - 0.191) |
| *Dicentrarchus labrax* | RZ | muscle | 1 | 349 | 346.6 | 0.151 | 0.000 | 0.000 | 0.000 | 0.000 |
| *Diplodus sargus* | IZ | muscle | 4 | 269 ± 21 (235 - 289) | 357.2 ± 86.9 (214.6 - 433.3) | 0.064 ± 0.01 (0.056 - 0.076) | 0.000 ± 0.000 (0.000 - 0.000) | 0.005 ± 0.007 (0.000 - 0.015) | 0.000 ± 0.000 (0.000 - 0.000) | 0.007 ± 0.003 (0.003 - 0.011) |
| *Diplodus sargus* | RZ | muscle | 39 | 245 ± 45 (173 - 375) | 308.7 ± 189.1 (101 - 969.4) | 0.061 ± 0.032 (0.035 - 0.177) | 0.015 ± 0.065 (0.000 - 0.382) | 0.023 ± 0.121 (0.000 - 0.757) | 0.044 ± 0.086 (0.000 - 0.509) | 0.004 ± 0.004 (0.000 - 0.020) |
| *Diplodus vulgaris* | IZ | muscle | 28 | 226 ± 20 (189-279) | 203.5 ± 59.0 (83.6 - 359.4) | 0.115 ± 0.051 (0.067 - 0.266) | 0.017 ± 0.068 (0.000 - 0.334) | 0.024 ± 0.070 (0.000 - 0.368) | 0.019 ± 0.038 (0.000 - 0.131) | 0.016 ± 0.023 (0.000 - 0.13) |
| *Diplodus vulgaris* | RZ | muscle | 53 | 213 ± 24 (149 - 249) | 164.0 ± 56.8 (46.9 - 283.9) | 0.102 ± 0.082 (0.032 - 0.475) | 0.000 ± 0.000 (0.000 - 0.000) | 0.025 ± 0.029 (0.000 - 0.125) | 0.019 ± 0.036 (0.000 - 0.197) | 0.019 ± 0.0260 (0.000 - 0.154) |
| *Engraulis encrasicolus* | IZ | muscle | 60 | 139 ± 8 (121-157) | 16.8 ± 2.9 (10.2 - 25.5) | 0.345 ± 0.091 (0.039 - 0.533) | 0.006 ± 0.025 (0.000 - 0.157) | 0.004 ± 0.002 (0.000 - 0.012) | 0.045 ± 0.051 (0.000 - 0.287) | 0.021 ± 0.012 (0.002 - 0.073) |
| *Engraulis encrasicolus* | RZ | muscle | 6 | 118 ± 11 (100 - 129) | 10.6 ± 2.7 (5.5 - 12.5) | 0.478 ± 0.210 (0.262 - 0.879) | 0.028 ± 0.023 (0.000 - 0.051) | 0.072 ± 0.100 (0.000 - 0.216) | 0.115 ± 0.105 (0.039 - 0.257) | 0.032 ± 0.021 (0.018 - 0.072) |
| *Galeus melastomus* | IZ | muscle | 32 | 459 ± 81 (323 - 578) | 283.4 ± 147.3 (81.8 - 545.7) | 0.118 ± 0.024 (0.081 - 0.198) | 0.007 ± 0.036 (0.000 - 0.201) | 0.053 ± 0.068 (0.005 - 0.305) | 0.191 ± 0.147 (0.054 - 0.754) | 0.003 ± 0.004 (0.000 - 0.019) |
| *Galeus melastomus* | RZ | muscle | 4 | 327 ± 31 (277 - 354) | 89.8 ± 23.1 (51.2 - 110.0) | 0.210 ± 0.098 (0.140 - 0.354) | 0.000 ± 0.000 (0.000 - 0.000) | 0.141 ± 0.104 (0.035 - 0.280) | 0.218 ± 0.03 (0.182 - 0.247) | 0.005 ± 0.002 (0.002 - 0.006) |
| *Helicolenus dactylopterus* | IZ | muscle | 37 | 187 ± 31 (114 - 248) | 109.6 ± 51.2 (22.2 - 264.9) | 0.096 ± 0.037 (0.055 - 0.276) | 0.006 ± 0.036 (0.000 - 0.221) | 0.086 ± 0.100 (0.006 - 0.435) | 0.127 ± 0.111 (0.000 - 0.647) | 0.004 ± 0.005 (0.000 - 0.026) |
| *Helicolenus dactylopterus* | RZ | muscle | 46 | 194 ± 33 (139-291) | 130.7 ± 72.2 (37.6 - 402) | 0.086 ± 0.025 (0.051 - 0.167) | 0.004 ± 0.019 (0.000 - 0.103) | 0.003 ± 0.003 (0.000 - 0.012) | 0.038 ± 0.031 (0.000 - 0.105) | 0.002 ± 0.002 (0.000 - 0.009) |
| *Liza aurata* | IZ | muscle | 12 | 281 ± 24 (241-330) | 193.3 ± 71.1 (111.7 - 388.8) | 0.094 ± 0.027 (0.064 - 0.155) | 0.010 ± 0.022 (0.000 - 0.060) | 0.001 ± 0.003 (0.000 - 0.008) | 0.021 ± 0.036 (0.000 - 0.114) | 0.003 ± 0.003 (0.000 - 0.011) |
| *Liza aurata* | RZ | muscle | 7 | 328 ± 25 (282 - 359) | 307.3 ± 65.6 (212.6 - 385.9) | 0.093 ± 0.054 (0.057 - 0.196) | 0.018 ± 0.025 (0.000 - 0.053) | 0.008 ± 0.01 (0.000 - 0.029) | 0.010 ± 0.017 (0.000 - 0.036) | 0.007 ± 0.005 (0.000 - 0.013) |
| *Merluccius merluccius* | IZ | liver | 48 | 364 ± 135 (233 - 795) | 471.6 ± 534.3 (97.7 - 2305.2) | 1.69 ± 0.545 (0.081 - 2.748) | 0.008 ± 0.027 (0.000 - 0.147) | 0.015 ± 0.019 (0.000 - 0.107) | 0.130 ± 0.229 (0.000 - 1.27) | 0.034 ± 0.030 (0.000 - 0.135) |
| *Merluccius merluccius* | IZ | muscle | 53 | 354 ± 132 (233 - 795) | 439.1 ± 518.4 (89.1 - 2305.2) | 0.145 ± 0.194 (0.067 - 1.151) | 0.031 ± 0.186 (0.000 - 1.314) | 0.01 ± 0.044 (0.000 - 0.308) | 0.018 ± 0.042 (0.000 - 0.229) | 0.002 ± 0.003 (0.000 - 0.022) |

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|---|---|---|---|---|---|---|---|---|---|---|
| *Merluccius merluccius* | RZ | muscle | 24 | 384 ± 54 (280 - 476) | 456.9 ±185.6 (147.3 – 858.4) | 0.082 ± 0.018 (0.054 - 0.121) | 0.023 ± 0.111 (0.000 - 0.544) | 0.026 ± 0.063 (0.000 - 0.293) | 0.011 ± 0.021 (0.000 - 0.057) | 0.000 ± 0.001 (0.000 - 0.002) |
| *Mugil cephalus* | IZ | muscle | 1 | 425 | 969.5 | 0.055 | 0.000 | 0.009 | 0.001 | 0.001 |
| *Mullus barbatus* | IZ | muscle | 77 | 171 ± 25 (121 - 230) | 57.7 ± 27.7 (11.7 – 140.7) | 0.159 ± 0.048 (0.05 - 0.332) | 0.005 ± 0.02 (0.000 - 0.101) | 0.014 ± 0.023 (0.000 - 0.192) | 0.076 ± 0.069 (0.000 - 0.35) | 0.006 ± 0.006 (0.002 - 0.046) |
| *Mullus barbatus* | RZ | muscle | 2 | 212 ± 28 (185 - 239) | 115.0 ± 45.6 (70.4 – 159.5) | 0.119 ± 0.025 (0.101 - 0.136) | 0.000 ± 0.000 (0.000 - 0.000) | 0.048 ± 0.006 (0.043 - 0.052) | 0.050 ± 0.071 (0.000 - 0.100) | 0.003 ± 0.001 (0.002 - 0.004) |
| *Mullus surmuletus* | IZ | muscle | 68 | 181 ± 29 (135 - 267) | 75.2 ± 41.1 (27.2 – 236.1) | 0.17 ± 0.058 (0.098 - 0.442) | 0.001 ± 0.01 (0.000 - 0.084) | 0.01 ± 0.018 (0.000 - 0.082) | 0.058 ± 0.051 (0.000 - 0.212) | 0.005 ± 0.003 (0.000 - 0.020) |
| *Mullus surmuletus* | RZ | muscle | 72 | 188 ± 30 (152 - 315) | 88.4 ± 57.9 (39.1 – 418.8) | 0.136 ± 0.041 (0.076 - 0.266) | 0.002 ± 0.010 (0.000 - 0.069) | 0.016 ± 0.011 (0.000 - 0.048) | 0.045 ± 0.047 (0.000 - 0.228) | 0.002 ± 0.003 (0.000 - 0.020) |
| *Pagellus bogaraveo* | IZ | muscle | 27 | 268 ± 19 (236 - 307) | 267.9 ± 63.6 (149.6 – 420.6) | 0.098 ± 0.032 (0.067 - 0.21) | 0.012 ± 0.060 (0.000 - 0.312) | 0.023 ± 0.037 (0.000 - 0.190) | 0.092 ± 0.109 (0.000 - 0.547) | 0.002 ± 0.003 (0.000 - 0.013) |
| *Pagellus bogaraveo* | RZ | muscle | 30 | 240 ± 10 (221 - 265) | 204.5 ± 24.4 (167.5 – 273.8) | 0.101 ± 0.061 (0.059 - 0.375) | 0.000 ± 0.000 (0.000 - 0.000) | 0.029 ± 0.035 (0.004 - 0.130) | 0.012 ± 0.035 (0.000 - 0.166) | 0.001 ± 0.001 (0.000 - 0.004) |
| *Pagellus erythrinus* | RZ | muscle | 6 | 172 ± 11 (154 - 188) | 68.3 ± 10.7 (55.1 – 83.1) | 0.112 ± 0.028 (0.076 - 0.148) | 0.000 ± 0.000 (0.000 - 0.000) | 0.082 ± 0.046 (0.042 - 0.162) | 0.116 ± 0.064 (0.071 - 0.208) | 0.004 ± 0.006 (0.000 - 0.012) |
| *Raja clavata* | IZ | muscle | 21 | 605 ± 116 (440 - 826) | 1493.4 ± 940.0 (431.4 – 3820.1) | 0.309 ± 0.279 (0.116 - 1.047) | 0.000 ± 0.000 (0.000 - 0.000) | 0.004 ± 0.009 (0.000 - 0.037) | 0.030 ± 0.044 (0.000 - 0.166) | 0.014 ± 0.045 (0.000 - 0.202) |
| *Raja clavata* | RZ | muscle | 5 | 749 ± 68 (669 - 849) | 2293.3 ± 650.1 (1567.7 – 3181.4) | 0.160 ± 0.023 (0.145 - 0.200) | 0.000 ± 0.000 (0.000 - 0.000) | 0.006 ± 0.011 (0.000 - 0.025) | 0.000 ± 0.000 (0.000 - 0.000) | 0.000 ± 0.001 (0.000 - 0.002) |
| *Sardina pilchardus* | IZ | muscle | 68 | 141 ± 11 (124 - 193) | 21.3 ± 5.9 (12.5 – 54.7) | 0.414 ± 0.098 (0.118 - 0.744) | 0.023 ± 0.024 (0.000 - 0.115) | 0.01 ± 0.004 (0.000 - 0.024) | 0.066 ± 0.039 (0.000 - 0.246) | 0.038 ± 0.023 (0.000 - 0.092) |
| *Sardinella aurita* | RZ | muscle | 2 | 240 ± 6 (234 - 246) | 101.3 ± 6.9 (94.5 – 108.0) | 0.829 ± 0.021 (0.814 - 0.844) | 0.000 ± 0.000 (0.000 - 0.000) | 0.021 ± 0.012 (0.012 - 0.029) | 0.053 ± 0.028 (0.033 - 0.073) | 0.02 ± 0.028 (0.000 - 0.04) |
| *Scorpaena porcus* | IZ | muscle | 39 | 187 ± 39 (129 - 262) | 155.2 ± 99.7 (45.1 – 392.6) | 0.117 ± 0.069 (0.059 - 0.468) | 0.003 ± 0.010 (0.000 - 0.050) | 0.04 ± 0.047 (0.000 - 0.188) | 0.096 ± 0.066 (0.000 - 0.377) | 0.003 ± 0.004 (0.000 - 0.014) |
| *Scorpaena porcus* | RZ | muscle | 71 | 160 ± 21 (119 - 218) | 83.6 ± 37.4 (33.7 – 201.7) | 0.119 ± 0.045 (0.063 - 0.350) | 0.006 ± 0.016 (0.000 - 0.089) | 0.041 ± 0.054 (0.000 - 0.325) | 0.107 ± 0.057 (0.035 - 0.252) | 0.004 ± 0.006 (0.000 - 0.025) |
| *Scyliorhinus canicula* | IZ | muscle | 63 | 417 ± 59 (244 - 509) | 245.1 ± 90.8 (39.4 – 400.4) | 0.272 ± 0.301 (0.121 - 1.794) | 0.002 ± 0.013 (0.000 - 0.084) | 0.024 ± 0.045 (0.000 - 0.321) | 0.112 ± 0.113 (0.000 - 0.435) | 0.009 ± 0.014 (0.000 - 0.087) |
| *Scyliorhinus canicula* | RZ | muscle | 82 | 370 ± 62 (244 - 500) | 183.6 ± 103.9 (41.3 – 485.4) | 0.226 ± 0.082 (0.000 - 0.726) | 0.013 ± 0.062 (0.000 - 0.521) | 0.039 ± 0.037 (0.000 - 0.248) | 0.117 ± 0.108 (0.000 - 0.678) | 0.009 ± 0.01 (0.000 - 0.057) |
| *Sparus aurata* | IZ | muscle | 24 | 252 ± 15 (232 - 306) | 233.9 ± 46.1 (183.6 – 419.6) | 0.089 ± 0.020 (0.058 - 0.127) | 0.022 ± 0.108 (0.000 - 0.527) | 0.003 ± 0.004 (0.000 - 0.014) | 0.014 ± 0.028 (0.000 - 0.093) | 0.001 ± 0.001 (0.000 - 0.004) |
| *Sparus aurata* | RZ | muscle | 4 | 261 ± 45 (223 - 336) | 264.7 ± 150.2 (150.2 – 519.6) | 0.070 ± 0.023 (0.036 - 0.090) | 0.000 ± 0.000 (0.000 - 0.000) | 0.007 ± 0.005 (0.000 - 0.012) | 0.059 ± 0.040 (0.000 - 0.088) | 0.002 ± 0.001 (0.000 - 0.002) |
| *Trachurus mediterraneus* | IZ | muscle | 8 | 331 ± 20 (307 - 366) | 283.4 ± 49.6 (238 - 393.6) | 0.085 ± 0.022 (0.049 - 0.111) | 0.000 ± 0.000 (0.000 - 0.000) | 0.007 ± 0.008 (0.000 - 0.021) | 0.045 ± 0.062 (0.000 - 0.177) | 0.002 ± 0.001 (0.001 - 0.003) |
| *Trachurus mediterraneus* | RZ | muscle | 30 | 272 ± 22 (247 - 318) | 165.2 ± 41.8 (115.3 – 256.8) | 0.102 ± 0.019 (0.057 - 0.135) | 0.024 ± 0.046 (0.000 - 0.170) | 0.02 ± 0.017 (0.002 - 0.083) | 0.061 ± 0.069 (0.000 - 0.35) | 0.002 ± 0.002 (0.000 - 0.006) |
| *Trachurus trachurus* | IZ | muscle | 34 | 337 ± 37 (281 - 425) | 352.1 ± 130.8 (180 – 659.1) | 0.096 ± 0.025 (0.071 - 0.197) | 0.016 ± 0.062 (0.000 - 0.326) | 0.008 ± 0.013 (0.000 - 0.056) | 0.037 ± 0.050 (0.000 - 0.195) | 0.001 ± 0.001 (0.000 - 0.004) |
| *Zeus faber* | IZ | muscle | 36 | 322 ± 96 (131 - 511) | 581.6 514.5 (38.1 – 2278.2) | 0.086 ± 0.040 (0.035 - 0.164) | 0.010 ± 0.040 (0.000 - 0.223) | 0.005 ± 0.016 (0.000 - 0.071) | 0.021 ± 0.050 (0.000 - 0.271) | 0.000 ± 0.000 (0.000 - 0.002) |

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|---|---|---|---|---|---|---|---|---|---|---|
| *Zeus faber* | RZ | muscle | 1 | 355 | 612.2 | 0.059 | 0.000 | 0.004 | 0.000 | 0.000 |

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668 **Table 2:** Mean TE concentrations ± standard deviation (in mg.kg$^{-1}$ wm) measured in fish liver from the zone (IZ) impacted by red mud and the reference (RZ)
669 zones. Minimum and maximum values are given in brackets.
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| Species | zone | Tissue | N | Total length (mm) | Total mass (g) | Al | As | Cd | Co | Cr | Hg |
|---|---|---|---|---|---|---|---|---|---|---|---|
| *Conger conger* | IZ | liver | 30 | 696 ± 157 (416 - 1020) | 729.7 ± 525.3 (94.5 – 2161.8) | 0.173 ± 0.131 (0.000 - 0.535) | 19.809 ± 9.831 (5.278 - 48.487) | 0.117 ± 0.082 (0.000 - 0.263) | 0.021 ± 0.007 (0.000 - 0.032) | 0.07 ± 0.084 (0.000 - 0.377) | 0.331 ± 0.203 (0.042 - 1.033) |
| *Conger conger* | RZ | liver | 21 | 691 ± 195 (334 - 1010) | 817.5 ± 672.6 (50.8- 2341.7) | 0.334 ± 0.458 (0.000 - 1.931) | 24.035 ± 14.403 (6.931 - 59.475) | 0.117 ± 0.127 (0.001 - 0.503) | 0.019 ± 0.009 (0.000 - 0.038) | 0.073 ± 0.208 (0.000 - 0.950) | 0.286 ± 0.211 (0.055 - 0.867) |
| *Merluccius merluccius* | IZ | liver | 48 | 364 ± 135 (233 - 795) | 471.6 ± 534.3 (97.7 - 2305.2) | 1.963 ± 2.563 (0.000 - 14.479) | 5.194 ± 1.859 (2.803 - 12.569) | 0.071 ± 0.101 (0.000 - 0.514) | 0.033 ± 0.016 (0.000 - 0.072) | 0.028 ± 0.044 (0.000 - 0.256) | 0.442 ± 0.729 (0.06 - 4.644) |
| *Merluccius merluccius* | RZ | liver | 23 | 388 ± 51 (284 - 476) | 469.7 ± 178.9 (147.3 – 858.4) | 0.922 ± 0.699 (0.000 - 3.13) | 3.961 ± 1.217 (2.413 - 8.010) | 0.06 ± 0.049 (0.012 - 0.208) | 0.025 ± 0.012 (0.006 - 0.053) | 0.026 ± 0.054 (0.000 - 0.165) | 0.261 ± 0.219 (0.064 - 0.972) |
| *Raja clavata* | IZ | liver | 21 | 605 ± 116 (440 - 826) | 1493.4 ± 940.0 (431.4 – 3820.1) | 0.819 ± 0.712 (0.000 - 2.885) | 33.368 ± 20.832 (13.88 - 81.629) | 0.291 ± 0.278 (0.001 - 1.255) | 0.04 ± 0.023 (0.000 - 0.090) | 0.005 ± 0.008 (0.000 - 0.029) | 0.666 ± 0.613 (0.261 - 2.476) |
| *Raja clavata* | RZ | liver | 5 | 749 ± 68 (669 - 849) | 2293.3 ± 650.1 (1567.7 – 3181.4) | 1.539 ± 1.146 (0.463 - 3.349) | 30.499 ± 4.233 (26.315 - 36.691) | 0.346 ± 0.230 (0.130 - 0.653) | 0.078 ± 0.025 (0.046 - 0.104) | 0.006 ± 0.013 (0.000 - 0.028) | 0.669 ± 0.521 (0.237 - 1.425) |
| *Scyliorhinus canicula* | IZ | liver | 60 | 417 ± 58 (244 - 509) | 244.1 ± 91.2 (39.4 – 400.4) | 1.873 ± 1.727 (0.238 - 7.033) | 29.800 ± 16.703 (3.827 - 81.501) | 0.202 ± 0.220 (0.000 - 0.971) | 0.059 ± 0.042 (0.003 - 0.186) | 0.036 ± 0.064 (0.000 - 0.393) | 1.129 ± 1.191 (0.121 - 6.146) |
| *Scyliorhinus canicula* | RZ | liver | 81 | 370 ± 62 (244 - 500) | 182.7 ± 104.2 (41.3 – 485.4) | 2.594 ± 2.362 (0.466 - 14.108) | 27.772 ± 12.341 (2.900 - 63.125) | 0.167 ± 0.149 (0.001 - 0.857) | 0.068 ± 0.034 (0.026 - 0.168) | 0.036 ± 0.037 (0.000 - 0.169) | 0.496 ± 0.383 (0.077 - 2.182) |

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**Table 2 (continued):** Mean TE concentrations ± standard deviation (in $mg.kg^{-1}$ wm) measured in fish liver from the zone (IZ) impacted by red mud and the reference (RZ) zones. Minimum and maximum values are given in brackets.

| Species | zone | tissue | N | Total length (mm) | Total mass (g) | Mn | Ni | Pb | Ti | V |
|---|---|---|---|---|---|---|---|---|---|---|
| *Conger conger* | IZ | liver | 30 | 696 ± 157 (416 - 1020) | 729.7 ± 525.3 (94.5 – 2161.8) | 0.934 ± 0.214 (0.280 - 1.361) | 0.022 ± 0.045 (0.000 - 0.186) | 0.069 ± 0.044 (0.000 - 0.169) | 0.021 ± 0.043 (0.000 - 0.151) | 0.096 ± 0.064 (0.003 - 0.331) |
| *Conger conger* | RZ | liver | 21 | 691 ± 195 (334 - 1010) | 817.5 ± 672.6 (50.8- 2341.7) | 1.119 ± 0.213 (0.768 - 1.553) | 0.000 ± 0.000 (0.000 - 0.000) | 0.102 ± 0.150 (0.008 - 0.657) | 0.055 ± 0.079 (0.000 - 0.208) | 0.131 ± 0.096 (0.006 - 0.368) |
| *Merluccius merluccius* | IZ | liver | 48 | 364 ± 135 (233 - 795) | 471.6 ± 534.3 (97.7 - 2305.2) | 1.69 ± 0.545 (0.081 - 2.748) | 0.008 ± 0.027 (0.000 - 0.147) | 0.015 ± 0.019 (0.000 - 0.107) | 0.130 ± 0.229 (0.000 - 1.27) | 0.034 ± 0.030 (0.000 - 0.135) |
| *Merluccius merluccius* | RZ | liver | 23 | 388 ± 51 (284 - 476) | 469.7 ± 178.9 (147.3 – 858.4) | 1.444 ± 0.383 (0.924 - 2.629) | 0.014 ± 0.037 (0.000 - 0.113) | 0.015 ± 0.025 (0.000 - 0.117) | 0.022 ± 0.035 (0.000 - 0.095) | 0.017 ± 0.012 (0.002 - 0.042) |
| *Raja clavata* | IZ | liver | 21 | 605 ± 116 (440 - 826) | 1493.4 ± 940.0 (431.4 – 3820.1) | 0.798 ± 0.336 (0.129 - 1.506) | 0.003 ± 0.014 (0.000 - 0.066) | 0.015 ± 0.012 (0.000 - 0.049) | 0.043 ± 0.062 (0.000 - 0.255) | 0.051 ± 0.049 (0.000 - 0.231) |
| *Raja clavata* | RZ | liver | 5 | 749 ± 68 (669 - 849) | 2293.3 ± 650.1 (1567.7 – 3181.4) | 0.859 ± 0.185 (0.571 - 1.023) | 0.013 ± 0.028 (0.000 - 0.063) | 0.019 ± 0.008 (0.008 - 0.027) | 0.036 ± 0.056 (0.000 - 0.128) | 0.081 ± 0.044 (0.04 - 0.151) |
| *Scyliorhinus canicula* | IZ | liver | 60 | 417 ± 58 (244 - 509) | 244.1 ± 91.2 (39.4 – 400.4) | 0.873 ± 0.435 (0.208 - 2.165) | 0.012 ± 0.042 (0.000 - 0.211) | 0.026 ± 0.017 (0.003 - 0.079) | 0.078 ± 0.084 (0.000 - 0.343) | 0.038 ± 0.034 (0.002 - 0.168) |
| *Scyliorhinus canicula* | RZ | liver | 81 | 370 ± 62 (244 - 500) | 182.7 ± 104.2 (41.3 – 485.4) | 1.147 ± 0.437 (0.488 - 3.143) | 0.011 ± 0.026 (0.000 - 0.112) | 0.029 ± 0.02 (0.007 - 0.151) | 0.074 ± 0.084 (0.000 - 0.442) | 0.129 ± 0.332 (0.007 - 2.421) |

**Table 3:** Speciation of arsenic in muscle of fish collected from the zone (IZ) impacted by red mud and the reference zone (RZ) (in $mg.kg^{-1}$ wm). Asi = inorganic arsenic (= As(III) because As(V) was not found in this study), MMA = monomethylarsonic acid, DMA = dimethylarsinic acid, AsB = arsenobetaine. Minimum and maximum values are given between brackets.

| Species | Zone | N | Length (mm) | mass (g) | Total As | Asi | MMA | DMA | AsB |
|---|---|---|---|---|---|---|---|---|---|
| *Diplodus sargus* | IZ | 3 | 264 ± 26 (235 - 285) | 335 ± 108.2 (214.6 - 424) | 6.23 ± 4.40 (2.34 - 11) | 0.033 ± 0.019 (0.013 - 0.048) | 0.001 ± 0.001 (0.000 - 0.002) | 0.004 ± 0.006 (0.000 - 0.011) | 5.69 ± 4.39 (1.81 - 10.5) |
| *Diplodus sargus* | RZ | 7 | 245 ± 32 (191 - 282) | 311.6 ± 121.5 (151 - 481.1) | 11.73 ± 6.60 (2.8 - 22.5) | 0.037 ± 0.015 (0.022 - 0.059) | 0.000 ± 0.000 (0.000 - 0.000) | 0.001 ± 0.002 (0.000 - 0.005) | 10.58 ± 6.71 (2.01 - 21.7) |
| *Diplodus vulgaris* | IZ | 5 | 227 ± 10 (213 - 239) | 204.4 ± 33.6 (165.8 - 240) | 4.44 ± 1.42 (2.64 - 6.41) | 0.06 ± 0.015 (0.037 - 0.073) | 0.000 ± 0.000 (0.000 - 0.000) | 0.005 ± 0.012 (0.000 - 0.026) | 4.35 ± 1.56 (2.43 - 6.52) |
| *Diplodus vulgaris* | RZ | 8 | 213 ± 9 (201 - 225) | 164.6 ± 19.3 (145.3 - 195.6) | 8.93 ± 4.08 (4.42 - 15.9) | 0.081 ± 0.009 (0.069 - 0.099) | 0.000 ± 0.001 (0.000 - 0.002) | 0.007 ± 0.002 (0.002 - 0.009) | 8.43 ± 4.23 (3.80 - 15.5) |
| *Helicolenus dactylopterus* | IZ | 5 | 186 ± 14 (168 - 201) | 108.7 ± 24.6 (76.8 - 135) | 5.52 ± 2.71 (2.46 - 9.18) | 0.021 ± 0.009 (0.014 - 0.033) | 0.000 ± 0.000 (0.000 - 0.000) | 0.001 ± 0.001 (0.000 - 0.002) | 4.33 ± 2.45 (1.79 - 7.61) |
| *Helicolenus dactylopterus* | RZ | 6 | 191 ± 21 (163 - 219) | 127 ± 36.3 (86.3 - 177.4) | 11.38 ± 7.30 (3.7 - 23.6) | 0.04 ± 0.029 (0.017 - 0.095) | 0.006 ± 0.01 (0.000 - 0.019) | 0.001 ± 0.001 (0.000 - 0.002) | 7.82 ± 5.74 (2.48 - 18.0) |
| *Pagellus bogaraveo* | IZ | 7 | 269 ± 11 (259 - 286) | 267.4 ± 38.8 (240.5 - 352.5) | 6.03 ± 2.31 (2.99 - 9.9) | 0.057 ± 0.009 (0.044 - 0.072) | 0.000 ± 0.001 (0.000 - 0.002) | 0.006 ± 0.003 (0.000 - 0.01) | 2.39 ± 0.71 (1.44 - 3.66) |
| *Pagellus bogaraveo* | RZ | 7 | 240 ± 5 (229 - 246) | 203.9 ± 12.4 (180.8 - 219.3) | 5.67 ± 1.55 (3.51 - 8.31) | 0.049 ± 0.02 (0.029 - 0.09) | 0.003 ± 0.007 (0.000 - 0.018) | 0.009 ± 0.007 (0.000 - 0.022) | 2.79 ± 1.31 (2.02 - 5.67) |
| *Pagellus erythrinus* | RZ | 2 | 172 ± 3 (170 - 174) | 68.3 ± 7.5 (63 - 73.6) | 4.73 ± 1.51 (3.67 - 5.8) | 0.041 ± 0.001 (0.04 - 0.042) | 0.006 ± 0.001 (0.005 - 0.007) | 0.013 ± 0.007 (0.008 - 0.018) | 4.58 ± 1.61 (3.44 - 5.71) |
| *Raja clavata* | IZ | 5 | 604 ± 72 (497 - 699) | 1474 ± 628.7 (677.6 - 2349.3) | 98.50 ± 19.94 (86 - 132) | 0.033 ± 0.015 (0.016 - 0.057) | 0.053 ± 0.021 (0.021 - 0.072) | 0.000 ± 0.000 (0.000 - 0.000) | 95.7 ± 19.9 (80.1 - 125) |
| *Raja clavata* | RZ | 2 | 744 ± 38 (717 - 771) | 2293.3 ± 286 (2091 - 2495.5) | 100.5 ± 27.58 (81 - 120) | 0.018 ± 0.003 (0.016 - 0.021) | 0.084 ± 0.045 (0.052 - 0.116) | 0.000 ± 0.000 (0.000 - 0.000) | 110 ± 26.8 (91.4 - 129) |
| *Sparus aurata* | IZ | 7 | 253 ± 10 (242 - 272) | 233.9 ± 27.1 (204.9 - 289.9) | 5.82 ± 3.29 (1.59 - 10.2) | 0.033 ± 0.026 (0.011 - 0.088) | 0.000 ± 0.000 (0.000 - 0.000) | 0.007 ± 0.003 (0.002 - 0.013) | 5.91 ± 3.92 (0.971 - 11.6) |
| *Sparus aurata* | RZ | 2 | 261 ± 26 (243 - 280) | 264.7 ± 99.5 (194.4 - 335.1) | 12.62 ± 10.02 (5.53 - 19.7) | 0.032 ± 0.002 (0.031 - 0.033) | 0.000 ± 0.000 (0.000 - 0.000) | 0.004 ± 0.005 (0.000 - 0.007) | 13.1 ± 10.1 (5.99 - 20.3) |

**Table 4:** Pearson correlation coefficients between TE concentrations and length in the muscle and the liver of fish collected in this study. Significant differences are highlighted by bold characters ($^{*}p < 0.05$, $^{**}p < 0.01$, $^{***}p < 0.001$)

| Species | Tissue | N IZ | N RZ | Al | As | Cd | Co | Cr | Hg | Mn | Ni | Pb | Ti | V |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| *Conger conger* | muscle | 31 | 23 | -0.057 | **0.56**$^{***}$ | -0.036 | -0.058 | **-0.337**$^{*}$ | **0.535**$^{***}$ | **-0.293**$^{*}$ | | **-0.42**$^{**}$ | **-0.469**$^{***}$ | **-0.290**$^{*}$ |
| *Coris julis* | muscle | 32 | 31 | -0.072 | **-0.258**$^{*}$ | -0.044 | **-0.347**$^{**}$ | -0.067 | 0.07 | **-0.252*** | -0.080 | -0.163 | -0.042 | **-0.458**$^{***}$ |
| *Diplodus vulgaris* | muscle | 28 | 53 | 0.096 | 0.075 | 0.116 | -0.162 | -0.041 | **0.738**$^{***}$ | -0.116 | **0.308**$^{**}$ | **-0.342**$^{**}$ | **-0.344**$^{**}$ | -0.125 |
| *Helicolenus dactylopterus* | muscle | 37 | 46 | **-0.251**$^{*}$ | -0.049 | -0.101 | -0.193 | 0.007 | **0.529**$^{***}$ | **-0.577**$^{***}$ | 0.021 | **-0.289**$^{**}$ | 0.133 | **-0.241**$^{*}$ |
| *Merluccius merluccius* | muscle | 53 | 24 | **-0.320**$^{**}$ | -0.084 | 0.027 | 0.041 | -0.016 | **0.851**$^{***}$ | -0.043 | -0.038 | **0.272**$^{*}$ | 0.147 | -0.077 |
| *Mullus surmuletus* | muscle | 68 | 72 | **-0.167**$^{*}$ | 0.136 | 0.077 | **0.181**$^{*}$ | 0.13 | **0.470**$^{***}$ | -0.004 | 0.052 | -0.034 | -0.043 | 0.122 |
| *Pagellus bogaraveo* | muscle | 27 | 30 | 0.004 | **0.270**$^{*}$ | 0.091 | **-0.274**$^{*}$ | -0.043 | 0.226 | 0.045 | 0.081 | 0.087 | 0.152 | -0.033 |
| *Scorpaena porcus* | muscle | 39 | 71 | **-0.363**$^{***}$ | 0.151 | -0.171 | -0.167 | -0.046 | **0.338**$^{***}$ | -0.163 | -0.112 | **-0.311**$^{**}$ | **-0.357**$^{***}$ | **-0.382**$^{***}$ |
| *Scyliorhinus canicula* | muscle | 63 | 82 | -0.120 | **0.250**$^{**}$ | 0.206 | 0.119 | **-0.352**$^{***}$ | **0.467**$^{***}$ | **0.166**$^{*}$ | **-0.167**$^{*}$ | -0.004 | **-0.343**$^{***}$ | 0.092 |
| *Conger conger* | liver | 30 | 21 | -0.197 | **0.472**$^{***}$ | 0.158 | **0.525**$^{***}$ | **-0.496**$^{***}$ | **0.444**$^{**}$ | 0.151 | -0.214 | -0.270 | -0.248 | **0.323**$^{*}$ |
| *Merluccius merluccius* | liver | 48 | 23 | **0.402**$^{**}$ | -0.186 | **0.583**$^{***}$ | **0.254*** | -0.173 | **0.748**$^{***}$ | **-0.449**$^{***}$ | -0.170 | **0.276**$^{*}$ | **0.472**$^{***}$ | **0.555**$^{***}$ |
| *Scyliorhinus canicula* | liver | 60 | 81 | **-0.182**$^{*}$ | **0.370**$^{***}$ | **0.337**$^{***}$ | **0.299**$^{***}$ | **-0.309**$^{***}$ | **0.434**$^{***}$ | **-0.260**$^{**}$ | **-0.211**$^{*}$ | 0.1 | **-0.314**$^{***}$ | **0.177**$^{*}$ |

Graphical abstract

