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Marc Bouchoucha, Rachida Chekri, A. Leufroy, Petru Jitaru, Sandrine Millour, et al.. Trace element contamination in fish impacted by bauxite red mud disposal in the Cassidaigne canyon (NW French Mediterranean). Science of the Total Environment, 2019, 690, pp.16 - 26. 10.1016/j.scitotenv.2019.06.474. hal-03487344

HAL Id: hal-03487344 https://hal.science/hal-03487344

Submitted on 21 Dec 2021

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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

1 Introduction

2 Hazards to human health stemming from exposure to individual contaminants or groups of 3 contaminants have emerged as a major concern in recent years (e.g. Copat et al., 2013; Damiano et 4 al., 2011; Miniero et al., 2014; Percin et al., 2011; Suarez-Serrano et al., 2010; Turkmen et al., 2009). 5 Among the wide range of toxic substances contaminating the marine environment, specific focus has 6 been given to trace elements (Castro-Gonzalez and Mendez-Armenta, 2008). Naturally present in the 7 Earth's crust, trace elements (TE) are also found in the marine environment (Bryan, 1971; Mason, 8 2013). However, human activities have considerably increased their spread into aquatic ecosystems 9 (Asante et al., 2010; Nriagu and Pacyna, 1988; Squadrone et al., 2016), especially those in marine 10 coastal areas (Ourgaud et al., 2018). TE are generally classified into two categories on the basis of 11 their metabolic role and their regulation by organisms. Essential TE (e.g. iron, copper and zinc) are 12 involved in metabolic processes, including functional and structural ones (Amiard et al., 1987). They 13 are generally efficiently regulated by marine animals, and present a narrow range of variation in their 14 tissues, except when ambient concentrations in seawater or food reach very high concentrations 15 (Amiard et al., 1987; Chapman, 1996; Vallee and Auld, 1990), or when they are involved in a specific 16 metabolic activity at a specific life stage (e.g. juvenile growth; Endo et al., 2008). On the contrary, 17 some TE (e.g. cadmium, lead and mercury) are not essential for marine life and their concentrations 18 in organisms depend mainly on environmental levels (Amiard et al., 1987). Whether essential or non-19 essential, TE become toxic above a certain threshold (Amiard et al., 1987).

20 From 1966 to 2015, the Gardanne alumina refinery discharged bauxite residue (called red mud)

21 through a pipe at 320 meters depth in the Cassidaigne Canyon in the northwest French

22 Mediterranean. This mud then spread along the canyon axis and on its lateral flanks to great depth

23 (>2000 m). A centimetric red layer of bauxite residue can now be observed along the canyon axis,

24 more than 50 km away from the pipe. The red mud contained high levels of TE, in particular titanium

25 (Ti), vanadium (V), aluminium (Al), and to a lesser extent arsenic (As) (Dauvin, 2010; Fontanier et al.,

26 2012). These elements are present in several mineral oxi-hydroxide phases (hematite, goethite, 27 ilmenite, rutile, anatase, brookite and kaolinite) that are not dissolved by the industrial process used 28 to extract alumina from bauxite ore at the Gardanne alumina refinery (Fontanier et al., 2012). Red 29 mud disposal into the Cassidaigne Canyon was expected to lower the biodiversity and abundance of 30 marine communities and to increase the potential uptake of bioavailable TE into the tissues of 31 marine organisms, their bioaccumulation through food webs and ultimately into human fish-32 consuming communities (Ellis et al., 1995; Gray et al., 2003; Metian et al., 2013; Swales et al., 1998; 33 Williams et al., 1999).

34 Since the 1960s, several environmental impact studies of the Gardanne alumina production activity 35 have been performed to document the effects of these disposals on biodiversity (Dauvin, 2010) but, 36 surprisingly, up to now the impact of red mud residues in the Cassidaigne Canyon on TE 37 contamination of fish and the risk related to fish consumption have been studied only twice. In 2004, 38 9 TE were analyzed in 28 samples from 12 fish species and in 2013, 27 TE were analyzed in 11 39 samples from 8 fish species (ANSES, 2016; Dauvin, 2010). These studies did not identify any 40 cumulated risk linked with fish consumption (Dauvin, 2010). However, their conclusions were limited: 41 (i) because the number of fish analyzed per species was very low; and (ii) because thresholds were 42 not compared with baseline values measured in a control zone, precluding any clear conclusion on 43 actual contamination. Indeed, for elements without regulatory thresholds such as Ti and V, a robust 44 impact assessment should include a comparison of trace metal concentrations in fish from the 45 impacted area and from an non-impacted area chosen as control. Moreover, As is one of the 46 elements present in the red muds discharged in the Cassidaigne Canyon. Arsenic can biomagnify in 47 food webs (Francesconi, 2010; Rahman et al., 2012), but this element exists in a number of chemical 48 forms in nature, all with very different toxicity characteristics, with inorganic species being the most 49 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 50 51 few data are available on As speciation in the Mediterranean context (see Fattorini et al., 2004;

52 Storelli et al., 2005; Storelli and Marcotrigiano, 2000). To our knowledge, As speciation in fish

53 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.

59

60 Materials and methods

61 Study area

62 The Cassidaigne Canyon is a short canyon (<50 km) located in the northern part of the western 63 Mediterranean Sea, east of the Gulf of Lions (Fig. 1). Its head borders Cassis Bay, only 8 km from the 64 coast, and presents in its deeper part two narrow passages located 3 km and 17 km, respectively, 65 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 66 67 tons (ANSES, 2016). Below 350 m, the entire seabed along the canyon axis was covered by the red 68 mud flow which spread more than 60 km from the pipe, with a significant proportion of the mud 69 being propagated at depths >2000 m (Dauvin, 2010). Today, this red mud also covers steep inclined 70 rock and is found underneath overhangs (Fabri et al., 2014). During strong north-eastern Mistral 71 wind events, especially during winter, quantities of this red mud could be transported northward, 72 from the canyon to the continental shelf (Dauvin, 2010). 73 To assess the impacts of red mud disposal on fish contamination, two zones were considered (Fig. 1).

74 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

'reference zone', RZ) was in the vicinity of the Stoechades Canyon. As water masses in this part of the
Mediterranean flow westward along the continental margin (Gatti et al., 2006), the Stoechades
Canyon is far from the influence of industrial discharge and thus not impacted by bauxite residues
(MC. Fabri, pers. com.). The head of the Stoechades Canyon is located in the Gulf of Hyères, less than
5 km from the coast, providing similar bathymetry and physico-chemical conditions that allow
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 between June 15th 2015 and July 27th 2015 by professional fishermen using different fishing gears 90 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.

Individual fish were stored on board and transferred to the laboratory in ice boxes where all fish
were measured (total length to the nearest mm) and weighed (total mass to the nearest 0.1 g)
before dissection. Specific measurements were performed in the muscle, the main edible tissue in
fish). White dorsal muscle was sampled, individually placed in polyethylene bags and kept frozen at 20°C until chemical analysis. For larger species, namely Conger conger, Merluccius merluccius,
Scyliorhinus canicula, Galeus melastomus and Raja clavata, livers were also collected when possible,
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 HNO₃ (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, 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 115 116 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 Pagellus 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 end of the sequence. The set criteria were as follows: calibration (r² > 0.995), blanks (values < limit of 139 140 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 $< \pm 2$) and duplicates 143 (acceptable if the relative standard deviation (RSD) \leq 20% when the mean value \geq 5 x LOQ or RSD \leq 144 40% when the mean value \geq 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).

All concentrations are given in mg.kg⁻¹ wm. When data were available in both muscle and liver, the
liver to muscle concentration ratios were calculated.

148

149 Statistical analyses

150 The statistical analysis of the results obtained on both matrices (muscle and liver) of fish was 151 performed only for species presenting a minimum sample size of 15 individuals per zone (IZ and RZ). 152 For these species, Pearson correlation tests were used to investigate the relationship between the TE 153 levels and fish length. Moreover, spatial variations in TE concentrations were analyzed according to 154 the zone for each matrix separately. To do this, PERMANOVAs were performed. They allow handling 155 complex, unbalanced multiple-factor designs and do not assume normal error distributions 156 (Anderson, 2001). Euclidean distance similarity matrices were generated and the factor 'sampling 157 area' was treated as fixed. Since it can affect contamination levels, fish length was considered as a 158 covariate. Data were log(X+1) transformed prior to statistical analyses. In each case, p-values were 159 calculated by 999 random permutations of residuals (Anderson, 2001). Concentrations below LOD 160 were set to 0 whereas concentrations below LOQ were set to LOD. Due to the large number of 161 species and TE analyzed, a large number of statistical analyses were performed simultaneously in this 162 study, thereby increasing the proportion of false positives. Therefore, a multiple testing correction 163 procedure was conducted to control the risk of reporting too many false positives and we used a 164 false discovery rate (FDR) method to adjust for the inflation of p-values (Benjamini and Hochberg, 165 1995). All data processing and statistical analyses were performed using R software (R Core Team, 166 2014) and PRIMER 6 software with the PERMANOVA add-on (Clarke and Warwick, 2001). The 167 significance level for the tests was consistently set at $\alpha = 0.05$.

168

169 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).

172 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*

174 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.

182 The trace element concentrations in the muscle and the liver of different fish species collected from

183 the two sampling areas are given in Table 2. In muscle, the distribution patterns in the TE

184 concentrations were as follows: As > Al > Hg > Mn > Ti > Pb > Cr > V > Ni > Cd > Co for IZ and As > Al >

185 Hg > Mn > Ti > Pb > Cr > V > Ni > Co > Cd for RZ. In the liver, the sequence was: As > Al > Mn > Hg > Cd

186 > Ti > Pb > Co > V > Cr > Ni for IZ and As > Al > Mn > Hg > Cd > V > Pb > Co > Ti > Cr > Ni for RZ.

187 Liver to muscle concentration ratios were consistently higher than 1 for Al, Cd, Co, Cr, Mn, Pb and V
188 and reached up to 360 for Cd in *M. merluccius* from RZ. Ratios were lower than 1 (*i.e.* higher

189 concentrations in muscle than in liver) for As and Hg. Due to the high number of values under LOD, it

190 was generally not possible to calculate those ratios for Ni. Finally, the liver to muscle concentration

191 ratios for Ti were below 1 for *C. conger* and *S. canicula* but over 1 for *M. merluccius* and *Raja clavata*.

192 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

194 found in the shark species *S. canicula* (p < 0.001) and the highest concentrations of Hg were found in

195 *H. dactylopterus* and *S. canicula* (p < 0.001). Considering livers, species-dependent patterns in TE

- 196 concentrations were less blurred and quite similar between sampling zones, with S. canicula
- 197 presenting the highest Al, As, Cd, Co and Hg concentrations, *M. merluccius* the highest Mn

198 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.

The relationships between fish length and concentrations differed between trace elements, species and tissues (Table 4). The vast majority of relationships, whether significant or not, were nonetheless negative, even for high concentrations of TE in the disposal. On the contrary, Hg was the only TE with a positive relationship for all species and tissues. Finally, As exhibited an intermediate pattern as positive relationships were observed for 8 species × tissue cases studied, 5 of which were significant (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 AI, 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

Very few studies have assessed the impact of the red mud disposal in the Cassidaigne Canyon oncontamination 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 lamiceli 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 mg.kg⁻¹ wm in teleosts, 1 mg.kg⁻¹ wm in sharks), confirming the concern raised previously regarding 237 238 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 of individuals with concentrations above the EU regulatory limits in fish muscle (0.3 mg.kg⁻¹ wm and 242 243 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

250 potential As poisoning through fish consumption should be further assessed through the specific 251 determination of the inorganic As content in fish tissues. Most of the As in the tissues of marine 252 animals is bound in organoarsenic forms (Neff, 1997) and considered of low toxicity (Hindmarsh, 253 2000). In this study, the main form of As found in fish was arsenobetaine which accounts for up to 254 100% of As in certain species (Table 3). This form is not toxic to humans (Neff, 1997). 255 In both sampling zones, our results clearly showed significant differences in the bioaccumulation of a 256 range of TE between the 26 selected species, suggesting a diversity response among fish (Table 1, 2). 257 Indeed, the accumulation of TE in fish does not directly reflect their simple concentration in the 258 ambient environment as it is affected by environmental factors, species biology and the physico-259 chemical properties of the contaminant (Cossa et al., 2012; Cresson et al., 2015). Thus, assessing a 260 site-specific contamination pattern requires good understanding of all these parameters although 261 the interpretation of the results remains complex because of the existence of interactions between 262 them. Notwithstanding some general trends could still be discerned. Some TE such as Hg are well-263 known for their biomagnification property, namely the trend for increasing concentrations along 264 food webs (Bryan et al., 1979). This process in particular is responsible for the high Hg concentrations 265 generally observed in high trophic level species such as marine apex predators (Endo et al., 2008) like 266 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-267 268 specific variation in mean trophic levels, ranging from 2.8 for L. aurata to 3.8 S. canicula (Cresson et 269 al., 2014b; Froese and Pauly, 2014; Stergiou and Karpouzi, 2001), could partially explain this 270 differences in the TE concentrations between these species. Moreover, an increasing number of 271 studies have documented relatively high levels of TE contamination in shelf-edge and deep species 272 (Cresson et al., 2014a; Hornung et al., 1993), especially when compared to coastal species 273 (Chouvelon et al., 2012). The 26 species selected here live at drastically different depths, from the 274 surface down to a few meters depth for C. julis, L. ramada and L. aurata, to several hundreds of 275 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 Atli, 2003; Dural et al., 2006; Fernandes et al., 2007; Long and Wang, 2005; Mormede and Davies, 314 315 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, AI 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.

361

362 Conclusion

363 The impact of the human activities on the levels of fish contamination by TE is not easy to assess in 364 situ because it is blurred by the interaction of biological effects (e.g. Bouchoucha et al., 2018; Cresson 365 et al., 2014a; Metian et al., 2013). A thorough sampling campaign and numerous analyses allowed 366 highlighting the significant contamination of fish species collected from the Cassidaigne Canyon area, 367 especially for Al and to a lesser extent for Ti and V. Although not statistically proved, it was also 368 shown that fish collected close to the alumina refinery pipe had higher Asi concentrations than those 369 from the reference area. In the next step, it would be interesting to assess human dietary exposure 370 to these metals through the consumption of fish from the Cassidaigne Canyon zone. This would allow 371 better evaluation of the health risk for humans consuming these fish. Moreover, the solubility of 372 many TE is highly redox-sensitive, thus the elemental concentrations in seawater change accordingly 373 (e.g. Morford and Emerson, 1999). Consequently, if the redox conditions change in the sites 374 contaminated by red mud, this would influence the solubility of TE, making them bioavailable to 375 marine organisms. Therefore, it seems essential to continue regular and long-term monitoring of 376 marine organisms from this area to prevent any further environmental deterioration and to assess 377 human exposure.

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379

380	Acknowledgements
381	This work received financial support from the Environment and Sustainable Development Ministry.
382	We are grateful to F. Ruchon and local fishermen for their help in fish sampling. Thanks are also due
383	to C. Tomasino, D. Louedoc, C. Ravel, A. Vaugier and F. Chavanon for their assistance during the
384	laboratory work.
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- 631 Figures
- 632
- **Figure 1:** Location of the sampling station and sampling zones. The star represents the Gardanne
- 634 alumina refinery pipeline outlet.
- 635



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)
 zones. Minimum and maximum values are given in brackets.

Species	zon e	Tissue	Ν	Total length (mm)	Total mass (g)	AI	As	Cd	Со	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)
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)
Dipiodus vulgaris	RZ	muscle	53	213 ± 24 (149 - 249)	(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)	(0.082 - 0.798)
encrasicolus	IZ	muscle	60	(121-157)	(10.2 - 25.5)	(0.030 ± 0.074) (0.000 - 0.363)	3.804 ± 0.759 (1.688 - 5.900)	(0.002 - 0.009)	(0.006 - 0.017)	(0.019 ± 0.018) (0.000 - 0.059)	(0.035 - 0.247)
encrasicolus	RZ	muscle	6	(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)
melastomus	IZ	muscle	32	459 ± 81 (323 - 578)	283.4 ± 147.3 (81.8 - 545.7)	(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)	(0.000 - 0.074)	(0.553 - 3.833)
melastomus	RZ	muscle	4	327 ± 31 (277 - 354)	89.8 ± 23.1 (51.2 - 110.0)	(0.812 - 2.829)	(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)	(0.799 - 1.089)
dactylopterus	IZ	muscle	37	(114 - 248)	(22.2 - 264.9)	(0.137-3.364)	(2.111 - 12.100)	(0.000 - 0.000)	(0.000 - 0.002)	(0.000 - 0.525)	(0.273 - 2.730)
dactylopterus	RZ	muscle	46	(139-291)	(37.6 - 402)	(0.000 - 1.839)	(2.690 - 25.008)	(0.000 ± 0.001)	(0.000 - 0.010)	(0.000 - 0.137)	(0.382 - 2.059)
Liza aurata	IZ	muscle	12	(241-330)	(111.7 - 388.8)	(0.000 - 0.217)	(0.285 - 0.960)	(0.000 ± 0.000) (0.000 ± 0.000)	(0.006 - 0.013)	(0.000 - 0.076)	(0.000 - 0.028)
Liza aurata	RZ	muscle	7	(282 - 359)	(212.6 - 385.9)	(0.000 - 0.218) 0.591 + 0.447	(0.832 - 2.230)	(0.000 ± 0.000) (0.000 + 0.000)	(0.000 - 0.018)	(0.000 - 0.088)	(0.013 - 0.047)
Liza ramada Marlucsius	IZ	muscle	13	(239 - 389)	(105.7 - 506.2)	(0.101 - 1.770)	(0.243 - 2.091)	(0.000 ± 0.000) (0.000 - 0.000)	(0.002 - 0.010)	(0.000 - 0.06)	(0.007 - 0.321)
merluccius	IZ	muscle	53	354 ± 132 (233 - 795)	439.1 ± 518.4 (89.1 - 2305.2)	(0.000 - 2.139)	(3.353 - 18.323)	(0.002 ± 0.007 (0.000 - 0.043)	(0.000 - 0.021)	(0.000 - 0.041)	(0.08 - 3.073)

Merluccius	D7	musclo	24	384 ± 54	456.9 ±185.6	0.131 ± 0.16	5.070 ± 2.194	0.000 ± 0.001	0.000 ± 0.000	0.006 ± 0.009	0.497 ± 0.39
merluccius	ΠĽ	muscle	24	(280 - 476)	(147.3 – 858.4)	(0.000 - 0.487)	(2.405 - 11.150)	(0.000 - 0.004)	(0.000 - 0.002)	(0.000 - 0.026)	(0.104 - 1.593)
Mugil	17	muscla	1	125	969 5	0 105	3 507	0.000	0.003	0.061	0 327
cephalus	12	muscie	1	425	505.5	0.105	5.557	0.000	0.005	0.001	0.527
Mullus	17	musclo	77	171 ± 25	57.7 ± 27.7	1.209 ± 1.434	24.775 ± 7.771	0.001 ± 0.002	0.008 ± 0.004	0.017 ± 0.02	0.46 ± 0.338
barbatus	12	muscle	//	(121 - 230)	(11.7 – 140.7)	(0.000 - 9.152)	(1.43 - 50.586)	(0.000 - 0.013)	(0.003 - 0.022)	(0.000 - 0.091)	(0.06 - 1.743)
Mullus	D7	mucclo	2	212 ± 28	115.0 ± 45.6	0.203 ± 0.163	13.222 ± 10.906	0.000 ± 0.000	0.007 ± 0.002	0.044 ± 0.037	0.217 ± 0.247
barbatus	ΠĽ	muscle	2	(185 - 239)	(70.4 – 159.5)	(0.088 - 0.318)	(5.510 - 20.933)	(0.000 - 0.000)	(0.005 - 0.008)	(0.018 - 0.07)	(0.042 - 0.391)
Mullus	17	musclo	60	181 ± 29	75.2 ± 41.1	1.294 ± 3.004	15.112 ± 5.732	0.000 ± 0.001	0.005 ± 0.002	0.014 ± 0.015	0.426 ± 0.51
surmuletus	١Z	muscle	00	(135 - 267)	(27.2 – 236.1)	(0.116 - 24.632)	(5.647 - 34.549)	(0.000 - 0.004)	(0.002 - 0.016)	(0.000 - 0.065)	(0.102 - 2.585)
Mullus	57	mucelo	72	188 ± 30	88.4 ± 57.9	0.268 ± 0.453	11.828 ± 5.166	0.000 ± 0.000	0.004 ± 0.003	0.014 ± 0.022	0.132 ± 0.098
surmuletus	KZ	muscle	72	(152 - 315)	(39.1 – 418.8)	(0.000 - 2.32)	(3.090 - 28.257)	(0.000 - 0.002)	(0.000 - 0.012)	(0.000 - 0.112)	(0.022 - 0.403)
Pagellus	17		27	268 ± 19	267.9 ± 63.6	0.328 ± 0.436	5.87 ± 2.535	0.001 ± 0.002	0.004 ± 0.003	0.009 ± 0.015	0.318 ± 0.174
bogaraveo	IZ	muscie	27	(236 - 307)	(149.6 – 420.6)	(0.000 - 2.083)	(2.250 - 11.900)	(0.000 - 0.010)	(0.000 - 0.012)	(0.000 - 0.066)	(0.092 - 1.041)
Pagellus			~~	240 ± 10	204.5 ± 24.4	0.203 ± 0.157	5.417 ± 1.138	0.001 ± 0.001	0.006 ± 0.002	0.017 ± 0.041	0.329 ± 0.194
bogaraveo	RZ	muscle	30	(221 - 265)	(167.5 – 273.8)	(0.000 - 0.671)	(3.212 - 7.746)	(0.000 - 0.002)	(0.003 - 0.014)	(0.000 - 0.220)	(0.183 - 0.995)
Pagellus				172 ± 11	68.3 ± 10.7	0.948 ± 0.191	5.06 ± 1.275	0.000 ± 0.000	0.002 ± 0.003	0.003 ± 0.007	0.171 ± 0.049
erythrinus	RZ	muscle	6	(154 - 188)	(55.1 – 83.1)	(0.743 - 1.27)	(3.640 - 6.350)	(0.000 - 0.000)	(0.000 - 0.006)	(0.000 - 0.016)	(0.091 - 0.234)
				605 ± 116	, 1493.4 ± 940.0	0.559 ± 1.143	76.051 ± 30.462	0.099 ± 0.300	0.013 ± 0.025	0.005 ± 0.009	1.185 ± 0.723
Raja clavata	IZ	muscle	21	(440 - 826)	(431.4 – 3820.1)	(0.000 - 4.987)	(1.404 - 119.085)	(0.001 - 1.325)	(0.000 - 0.088)	(0.000 - 0.029)	(0.156 - 3.765)
			_	, 749 ± 68	2293.3 ± 650.1	0.080 ± 0.129	89.171 ± 22.568	0.001 ± 0.001	0.002 ± 0.001	0.015 ± 0.014	1.429 ± 0.74
Raja clavata	RZ	muscle	5	(669 - 849)	(1567.7 – 3181.4)	(0.000 - 0.296)	(68.400 - 119.268)	(0.001 - 0.002)	(0.000 - 0.003)	(0.000 - 0.028)	(0.739 - 2.323)
Sardina				141 ± 11	21.3 ± 5.9	0.393 ± 0.968	5.805 ± 1.341	0.001 ± 0.002	0.011 ± 0.002	0.009 ± 0.014	0.1 ± 0.042
pilchardus	IZ	muscle	68	(124 - 193)	(12.5 - 54.7)	(0.000 - 7.817)	(4.028 - 12.264)	(0.000 - 0.007)	(0.007 - 0.016)	(0.000 - 0.073)	(0.046 - 0.334)
Sardinella			_	240 ± 6	101.3 ± 6.9	0.068 ± 0.096	6.230 ± 1.061	0.001 ± 0.000	0.003 ± 0.004	0.007 ± 0.009	0.103 ± 0.000
aurita	RZ	muscle	2	(234 - 246)	(94.5 - 108.0)	(0.000 - 0.136)	(5.480 - 6.980)	(0.001 - 0.001)	(0.000 - 0.006)	(0.000 - 0.013)	(0.103 - 0.103)
Scorpaena				187 ± 39	155.2 ± 99.7	0.808 ± 0.861	4.190 ± 2.439	0.001 ± 0.001	0.001 ± 0.001	0.013 ± 0.02	0.151 ± 0.049
porcus	IZ	muscle	39	(129 - 262)	(45.1 – 392.6)	(0.000-3.689)	(0.735 - 11.700)	(0.000 - 0.006)	(0.000 - 0.005)	(0.000 - 0.096)	(0.072 - 0.307)
Scornaena				160 + 21	83.6 + 37.4	0.778 ± 0.745	4.826 + 3.0590	0.001 + 0.001	0.003 ± 0.004	0.012 ± 0.026	0.171 ± 0.061
norcus	RZ	muscle	71	(119 - 218)	(33.7 - 201.7)	(0.108 - 3.74)	(0.577 - 12.600)	(0.000 - 0.006)	(0.000 - 0.015)	(0.000 - 0.153)	(0.072 - 0.361)
Scyliorhinus				417 + 59	245 1 + 90 8	1 259 + 1 727	37 735 + 17 946	0.065 + 0.221	0.012 ± 0.026	0.032 ± 0.026	1 613 + 1 335
canicula	IZ	muscle	63	(244 - 509)	(39.4 - 400.4)	(0 196 - 11 497)	(15 030 - 112 802)	(0.000 ± 0.221)	(0.012 ± 0.020)	(0.000 ± 0.020)	(0 233 - 8 955)
Scyliorhinus				370 + 62	183 6 + 103 9	1 299 + 0 995	41 341 + 19 253	(0.000 + 0.003)	(0.000 + 0.005)	$(0.000 \ 0.144)$ 0.042 + 0.034	0.845 + 0.309
canicula	RZ	muscle	82	(244 - 500)	(A1.3 - A85.4)	(0.000 - 4.815)	(0.020 - 118.615)	(0.002 ± 0.005)	(0.007 ± 0.003)	(0.042 ± 0.054)	(0.049 ± 0.303)
cuniculu				(244 - 300) 252 + 15	222 0 + 46 1	(0.000 - 4.013) 0.021 + 0.075	(0.020 - 110.013) 4 816 + 2 0760	(0.000 - 0.020)	(0.000 - 0.034)	(0.000 - 0.234)	(0.000 - 1.002) 0.15 + 0.087
Sparus aurata	IZ	muscle	24	(222 - 206)	(182.6 - 410.6)	(0.001 ± 0.075)	(1 262 - 10 010)	(0.000 ± 0.000)	(0.004 ± 0.002)	(0.000 ± 0.010)	(0.077 - 0.411)
				(232 - 300)	(183.0 - 419.0)	(0.000 - 0.234)	(1.203 - 10.910)	(0.000 - 0.002)	(0.002 - 0.003)	(0.000 + 0.003)	(0.077 - 0.411)
Sparus aurata	RZ	muscle	4	201 ± 45 (222 - 226)	204.7 ± 150.2	(0.147 ± 0.055)	11.940 ± 0.455	(0.000 ± 0.000)	(0.003 ± 0.001)	(0.009 ± 0.011)	(0.195 ± 0.100)
Trachurus				(225 - 550)	(130.2 - 319.0)	(0.117 - 0.109)	(4.940 - 25.710)	(0.000 - 0.000)	(0.002 - 0.003)	(0.000 - 0.024)	(0.050 - 0.421)
moditorranous	IZ	muscle	8	331 ± 20	283.4 ± 49.0	(0.0441 ± 0.420)	1.595 ± 1.122	0.002 ± 0.004	(0.005 ± 0.001)	(0.023 ± 0.024)	(0.311 ± 0.133)
Trachura				(307 - 300)	(238 - 393.0)	(0.000 - 1.081)	(0.549 - 3.889)	(0.000 - 0.011)	(0.003 - 0.008)	(0.000 - 0.074)	(0.342 - 0.741)
Trachurus	RZ	muscle	30	2/2±22	165.2 ± 41.8	0.298 ± 0.260	1.815 ± 1.113	0.001 ± 0.002	0.007 ± 0.005	0.011 ± 0.019	$0.3/8 \pm 0.135$
mealterraneus				(247 - 318)	(115.3 - 256.8)	(0.000 - 0.921)	(0.735 - 4.63)	(0.000 - 0.009)	(0.000 - 0.026)	(0.000 - 0.071)	(0.194 - 0.697)
iracnurus	IZ	muscle	34	33/±3/	352.1 ± 130.8	$0.1/6 \pm 0.110$	5.429 ± 2.0460	0.002 ± 0.005	0.008 ± 0.004	0.015 ± 0.03	0.947 ± 0.334
trachurus				(281 - 425)	(180 - 659.1)	(0.000 - 0.427)	(1.4/8 - 10.51)	(0.000 - 0.029)	(0.000 - 0.022)	(0.000 - 0.1/3)	(0.399 - 1.88)
Zeus faber	IZ	muscle	36	322 ± 96	581.6 514.5	0.142 ± 0.340	0.619 ± 0.308	0.001 ± 0.004	0.001 ± 0.001	0.013 ± 0.03	0.569 ± 0.496
,				(131 - 511)	(38.1 – 2278.2)	(0.000 - 2.030)	(0.302 - 1.406)	(0.000 - 0.019)	(0.000 - 0.004)	(0.000 - 0.133)	(0.046 - 2.251)

	Zeus faber	RZ	muscle	1	355	612.2	0.130	0.469	0.000	0.002	0.101	0.25
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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
 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
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	817.5 ± 672.6	0.515 ± 0.383	0.000 ± 0.000	0.006 ± 0.008	0.070 ± 0.115	0.004 ± 0.004
Coris julis	IZ	muscle	32	128 ± 1.9	20.9 ± 12.5	0.182 ± 0.058	0.007 ± 0.024	0.025 ± 0.019	0.059 ± 0.06	(0.030 ± 0.017) (0.000 - 0.090)
Coris julis	RZ	muscle	31	(103 - 212) 125 ± 14	(5.0 + 5.3) 17.0 ± 5.3 (5.1 + 20.4)	(0.037 ± 0.304) 0.196 ± 0.074	0.003 ± 0.011	0.014 ± 0.012	(0.000 ± 0.234) 0.114 ± 0.127 (0.000 ± 0.512)	(0.000 ± 0.000) 0.048 ± 0.043 (0.007 + 0.101)
Dicentrarchus	RZ	muscle	1	(86 - 147) 349	(5.1 - 29.4) 346.6	0.151	0.000	0.000 - 0.043)	0.000 - 0.513)	0.000
Diplodus	IZ	muscle	4	269 ± 21	357.2 ± 86.9	0.064 ± 0.01	0.000 ± 0.000	0.005 ± 0.007	0.000 ± 0.000	0.007 ± 0.003
Diplodus	RZ	muscle	39	(235 - 289) 245 ± 45	(214.6 - 433.3) 308.7 ± 189.1	(0.056 - 0.076) 0.061 ± 0.032	(0.000 - 0.000) 0.015 ± 0.065	(0.000 - 0.015) 0.023 ± 0.121	(0.000 - 0.000) 0.044 ± 0.086	(0.003 - 0.011) 0.004 ± 0.004
Diplodus	IZ	muscle	28	(1/3 - 3/5) 226 ± 20	(101 - 969.4) 203.5 ± 59.0	(0.035 - 0.177) 0.115 ± 0.051	(0.000 - 0.382) 0.017 ± 0.068	(0.000 - 0.757) 0.024 ± 0.070	(0.000 - 0.509) 0.019 ± 0.038	(0.000 - 0.020) 0.016 ± 0.023
Diplodus	RZ	muscle	53	(189-279) 213 ± 24	(83.6 - 359.4) 164.0 ± 56.8	(0.067 - 0.266) 0.102 ± 0.082	(0.000 - 0.334) 0.000 ± 0.000	(0.000 - 0.368) 0.025 ± 0.029	(0.000 - 0.131) 0.019 ± 0.036	(0.000 - 0.13) 0.019 ± 0.0260
vulgaris Engraulis	IZ	muscle	60	(149 - 249) 139 ± 8	(46.9 - 283.9) 16.8 ± 2.9	(0.032 - 0.475) 0.345 ± 0.091	(0.000 - 0.000) 0.006 ± 0.025	(0.000 - 0.125) 0.004 ± 0.002	(0.000 - 0.197) 0.045 ± 0.051	(0.000 - 0.154) 0.021 ± 0.012
encrasicolus Engraulis	R7	muscle	6	(121-157) 118 ± 11	(10.2 - 25.5) 10.6 ± 2.7	(0.039 - 0.533) 0.478 ± 0.210	(0.000 - 0.157) 0.028 ± 0.023	(0.000 - 0.012) 0.072 ± 0.100	(0.000 - 0.287) 0.115 ± 0.105	(0.002 - 0.073) 0.032 ± 0.021
encrasicolus Galeus	17	muscle	27	(100 - 129) 459 ± 81	(5.5 - 12.5) 283.4 ± 147.3	(0.262 - 0.879) 0.118 ± 0.024	(0.000 - 0.051) 0.007 ± 0.036	(0.000 - 0.216) 0.053 ± 0.068	(0.039 - 0.257) 0.191 ± 0.147	(0.018 - 0.072) 0.003 ± 0.004
melastomus Galeus	12	inuscie	52	(323 - 578) 327 ± 31	(81.8 - 545.7) 89.8 ± 23.1	(0.081 - 0.198) 0.210 ± 0.098	(0.000 - 0.201) 0.000 ± 0.000	(0.005 - 0.305) 0.141 ± 0.104	(0.054 - 0.754) 0.218 ± 0.03	(0.000 - 0.019) 0.005 ± 0.002
melastomus Helicolenus	RZ	muscle	4	(277 - 354) 187 ± 31	(51.2 - 110.0) 109.6 ± 51.2	(0.140 - 0.354) 0.096 ± 0.037	(0.000 - 0.000) 0.006 ± 0.036	(0.035 - 0.280) 0.086 ± 0.100	(0.182 - 0.247) 0.127 ± 0.111	(0.002 - 0.006) 0.004 ± 0.005
dactylopterus Helicolenus	IZ	muscle	37	(114 - 248)	(22.2 - 264.9)	(0.055 - 0.276) 0.086 + 0.025	(0.000 - 0.221) 0.004 + 0.019	(0.006 - 0.435)	(0.000 - 0.647)	(0.000 - 0.026)
dactylopterus	RZ	muscle	46	(139-291)	(37.6 - 402)	(0.051 - 0.167)	(0.000 - 0.103) (0.000 - 0.103)	(0.000 - 0.012) 0.001 + 0.003	(0.000 - 0.105) (0.000 - 0.105)	(0.000 - 0.009) (0.000 + 0.009)
Liza aurata	IZ	muscle	12	(241-330)	(111.7 - 388.8)	(0.064 - 0.155)	(0.000 - 0.060)	(0.000 - 0.008)	(0.000 - 0.114)	(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)	(0.000 - 0.053)	(0.000 ± 0.01 (0.000 - 0.029)	(0.010 ± 0.017) (0.000 - 0.036)	(0.007 ± 0.005)
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)

Merluccius	D7	muselo	24	384 ± 54	456.9 ±185.6	0.082 ± 0.018	0.023 ± 0.111	0.026 ± 0.063	0.011 ± 0.021	0.000 ± 0.001
merluccius	RZ	muscie	24	(280 - 476)	(147.3 – 858.4)	(0.054 - 0.121)	(0.000 - 0.544)	(0.000 - 0.293)	(0.000 - 0.057)	(0.000 - 0.002)
Mugil	17	musclo	1	125	060 5	0.055	0.000	0.000	0.001	0.001
cephalus	12	muscie	1	425	505.5	0.055	0.000	0.005	0.001	0.001
Mullus	17	musclo	77	171 ± 25	57.7 ± 27.7	0.159 ± 0.048	0.005 ± 0.02	0.014 ± 0.023	0.076 ± 0.069	0.006 ± 0.006
barbatus	12	muscle	//	(121 - 230)	(11.7 – 140.7)	(0.05 - 0.332)	(0.000 - 0.101)	(0.000 - 0.192)	(0.000 - 0.35)	(0.002 - 0.046)
Mullus	D7	musclo	r	212 ± 28	115.0 ± 45.6	0.119 ± 0.025	0.000 ± 0.000	0.048 ± 0.006	0.050 ± 0.071	0.003 ± 0.001
barbatus	RΖ	muscle	Z	(185 - 239)	(70.4 – 159.5)	(0.101 - 0.136)	(0.000 - 0.000)	(0.043 - 0.052)	(0.000 - 0.100)	(0.002 - 0.004)
Mullus	17	musclo	60	181 ± 29	75.2 ± 41.1	0.17 ± 0.058	0.001 ± 0.01	0.01 ± 0.018	0.058 ± 0.051	0.005 ± 0.003
surmuletus	١٢	muscle	00	(135 - 267)	(27.2 – 236.1)	(0.098 - 0.442)	(0.000 - 0.084)	(0.000 - 0.082)	(0.000 - 0.212)	(0.000 - 0.020)
Mullus	07		70	188 ± 30	88.4 ± 57.9	0.136 ± 0.041	0.002 ± 0.010	0.016 ± 0.011	0.045 ± 0.047	0.002 ± 0.003
surmuletus	KZ	muscle	72	(152 - 315)	(39.1 – 418.8)	(0.076 - 0.266)	(0.000 - 0.069)	(0.000 - 0.048)	(0.000 - 0.228)	(0.000 - 0.020)
Pagellus	17		27	268 ± 19	267.9 ± 63.6	0.098 ± 0.032	0.012 ± 0.060	0.023 ± 0.037	0.092 ± 0.109	0.002 ± 0.003
bogaraveo	IZ	muscle	27	(236 - 307)	(149.6 – 420.6)	(0.067 - 0.21)	(0.000 - 0.312)	(0.000 - 0.190)	(0.000 - 0.547)	(0.000 - 0.013)
Pagellus	57		20	240 ± 10	204.5 ± 24.4	0.101 ± 0.061	0.000 ± 0.000	0.029 ± 0.035	0.012 ± 0.035	0.001 ± 0.001
bogaraveo	RZ	muscle	30	(221 - 265)	(167.5 – 273.8)	(0.059 - 0.375)	(0.000 - 0.000)	(0.004 - 0.130)	(0.000 - 0.166)	(0.000 - 0.004)
Pagellus			~	172 ± 11	68.3 ± 10.7	0.112 ± 0.028	0.000 ± 0.000	0.082 ± 0.046	0.116 ± 0.064	0.004 ± 0.006
erythrinus	RZ	muscle	6	(154 - 188)	(55.1 – 83.1)	(0.076 - 0.148)	(0.000 - 0.000)	(0.042 - 0.162)	(0.071 - 0.208)	(0.000 - 0.012)
				605 ± 116	1493.4 ± 940.0	0.309 ± 0.279	0.000 ± 0.000	0.004 ± 0.009	0.030 ± 0.044	0.014 ± 0.045
Raja clavata	IZ	muscle	21	(440 - 826)	(431.4 - 3820.1)	(0.116 - 1.047)	(0.000 - 0.000)	(0.000 - 0.037)	(0.000 - 0.166)	(0.000 - 0.202)
			_	749 ± 68	2293.3 ± 650.1	0.160 ± 0.023	0.000 ± 0.000	0.006 ± 0.011	0.000 ± 0.000	0.000 ± 0.001
Raja clavata	RZ	muscle	5	(669 - 849)	(1567.7 – 3181.4)	(0.145 - 0.200)	(0.000 - 0.000)	(0.000 - 0.025)	(0.000 - 0.000)	(0.000 - 0.002)
Sardina				141 ± 11	21.3 ± 5.9	0.414 ± 0.098	0.023 ± 0.024	0.01 ± 0.004	0.066 ± 0.039	0.038 ± 0.023
pilchardus	IZ	muscle	68	(124 - 193)	(12.5 - 54.7)	(0.118 - 0.744)	(0.000 - 0.115)	(0.000 - 0.024)	(0.000 - 0.246)	(0.000 - 0.092)
Sardinella				240 ± 6	101.3 ± 6.9	0.829 ± 0.021	0.000 ± 0.000	0.021 ± 0.012	0.053 ± 0.028	0.02 ± 0.028
aurita	RZ	muscle	2	(234 - 246)	(94.5 - 108.0)	(0.814 - 0.844)	(0.000 - 0.000)	(0.012 - 0.029)	(0.033 - 0.073)	(0.000 - 0.04)
Scorpaena				187 ± 39	155.2 ± 99.7	0.117 ± 0.069	0.003 ± 0.010	0.04 ± 0.047	0.096 ± 0.066	0.003 ± 0.004
norcus	IZ	muscle	39	(129 - 262)	(45.1 - 392.6)	(0.059 - 0.468)	(0,000 - 0,050)	(0,000 - 0,188)	(0,000 - 0,377)	(0.000 - 0.014)
Scornaena				160 + 21	836+374	0.119 ± 0.045	0.006 ± 0.016	0.041 + 0.054	(0.000 - 0.077) 0 107 + 0 057	0.004 + 0.006
norcus	RZ	muscle	71	(119 - 218)	(33 7 - 201 7)	(0.063 - 0.350)	(0,000 - 0,089)	(0.000 - 0.325)	(0.035 - 0.252)	(0.000 - 0.025)
Scyliorhinus				417 + 59	245 1 + 90 8	$(0.003 \ 0.000)$	0.002 ± 0.013	(0.000 - 0.025) 0.024 + 0.045	(0.000 - 0.202) 0.112 + 0.113	0.009 ± 0.014
canicula	IZ	muscle	63	(244 - 509)	(39.4 - 400.4)	(0 121 - 1 79 <i>1</i>)	(0.002 ± 0.013)	(0.024 ± 0.043)	(0.000 - 0.435)	(0.000 ± 0.017)
Scyliorhinus				370 + 62	183 6 + 103 9	$(0.121 \ 1.754)$ 0.226 + 0.082	$(0.000 \ 0.004)$ 0.013 + 0.062	$(0.000 \ 0.021)$ 0.039 + 0.037	$(0.000 \ 0.400)$	(0.000 + 0.007)
canicula	RZ	muscle	82	(244 - 500)	(11.3 - 185.4)	(0.000 ± 0.002)	(0.013 ± 0.002)	(0.000 ± 0.007)	(0.000 - 0.678)	(0.000 ± 0.01)
cumculu				252 + 15	222 0 + 46 1	(0.000 - 0.720)	(0.000 - 0.021) 0.022 + 0.108	(0.000 - 0.248)	(0.000 - 0.078)	(0.000 - 0.057)
Sparus aurata	IZ	muscle	24	(222 - 206)	(182.6 - 410.6)	(0.059 ± 0.020)	(0.022 ± 0.108)	(0.003 ± 0.004)	(0.014 ± 0.028)	(0.001 ± 0.001)
				(232 - 300) 261 + 45	(183.0 - 413.0) 264 7 + 150 2	(0.038 - 0.127)	(0.000 - 0.327)	(0.000 - 0.014)	(0.000 - 0.093)	(0.000 - 0.004) 0.002 + 0.001
Sparus aurata	RZ	muscle	4	(201 ± 45	(150.2 510.6)	(0.070 ± 0.023)	(0.000 ± 0.000)	(0.007 ± 0.003)	(0.009 ± 0.040)	(0.002 ± 0.001)
Trachuruc				223 - 330)	(130.2 - 313.0)	(0.030 - 0.030)	(0.000 - 0.000)	(0.000 - 0.012)	(0.000 - 0.088)	(0.000 - 0.002)
moditorranous	IZ	muscle	8	(207 266)	205.4 ± 49.0	(0.065 ± 0.022)	(0.000 ± 0.000)	(0.007 ± 0.008)	(0.045 ± 0.002)	(0.002 ± 0.001)
Trachuruc				(307 - 300)	(238 - 393.0)	(0.049 - 0.111)	(0.000 - 0.000)	(0.000 - 0.021)	(0.000 - 0.177)	(0.001 - 0.003)
Tructiurus	RZ	muscle	30	$2/2 \pm 22$	105.2 ± 41.8	0.102 ± 0.019	(0.024 ± 0.046)	0.02 ± 0.017	(0.001 ± 0.009)	0.002 ± 0.002
Trachuraneus				(247 - 318)	(115.5 - 250.8)	(0.057 - 0.135)	(0.000 - 0.170)	(0.002 - 0.083)	(0.000 - 0.35)	
Trachurus	IZ	muscle	34	33/±3/	352.1 ± 130.8	0.090 ± 0.025	0.010 ± 0.062	0.008 ± 0.013	0.037 ± 0.050	0.001 ± 0.001
tracnurus				(281 - 425)	(180 - 659.1)	(0.071 - 0.197)	(0.000 - 0.326)	(0.000 - 0.056)	(0.000 - 0.195)	(0.000 - 0.004)
Zeus faber	IZ	muscle	36	322 ± 96	581.6 514.5	0.086 ± 0.040	0.010 ± 0.040	0.005 ± 0.016	0.021 ± 0.050	0.000 ± 0.000
				(131 - 511)	(38.1 – 2278.2)	(0.035 - 0.164)	(0.000 - 0.223)	(0.000 - 0.071)	(0.000 - 0.271)	(0.000 - 0.002)

Zeus faber	RZ	muscle	1	355	612.2	0.059	0.000	0.004	0.000	0.000
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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)
 zones. Minimum and maximum values are given in brackets.

Species	zon e	Tissue	N	Total length (mm)	Total mass (g)	AI	As	Cd	Со	Cr	Hg
Congor congor	17	livor	20	696 ± 157	729.7 ± 525.3	0.173 ± 0.131	19.809 ± 9.831	0.117 ± 0.082	0.021 ± 0.007	0.07 ± 0.084	0.331 ± 0.203
conger conger	IZ	liver	30	(416 - 1020)	(94.5 – 2161.8)	(0.000 - 0.535)	(5.278 - 48.487)	(0.000 - 0.263)	(0.000 - 0.032)	(0.000 - 0.377)	(0.042 - 1.033)
6	57	P	24	691 ± 195	817.5 ± 672.6	0.334 ± 0.458	24.035 ± 14.403	0.117 ± 0.127	0.019 ± 0.009	0.073 ± 0.208	0.286 ± 0.211
Conger conger	KZ	liver	21	(334 - 1010)	(50.8-2341.7)	(0.000 - 1.931)	(6.931 - 59.475)	(0.001 - 0.503)	(0.000 - 0.038)	(0.000 - 0.950)	(0.055 - 0.867)
Merluccius	17	12	40	364 ± 135	471.6 ± 534.3	1.963 ± 2.563	5.194 ± 1.859	0.071 ± 0.101	0.033 ± 0.016	0.028 ± 0.044	0.442 ± 0.729
merluccius	IZ	liver	48	(233 - 795)	(97.7 - 2305.2)	(0.000 - 14.479)	(2.803 - 12.569)	(0.000 - 0.514)	(0.000 - 0.072)	(0.000 - 0.256)	(0.06 - 4.644)
Merluccius	57	1	22	388 ± 51	469.7 ± 178.9	0.922 ± 0.699	3.961 ± 1.217	0.06 ± 0.049	0.025 ± 0.012	0.026 ± 0.054	0.261 ± 0.219
merluccius	KZ	liver	23	(284 - 476)	(147.3 – 858.4)	(0.000 - 3.13)	(2.413 - 8.010)	(0.012 - 0.208)	(0.006 - 0.053)	(0.000 - 0.165)	(0.064 - 0.972)
Daia davata	17	1	21	605 ± 116	1493.4 ± 940.0	0.819 ± 0.712	33.368 ± 20.832	0.291 ± 0.278	0.04 ± 0.023	0.005 ± 0.008	0.666 ± 0.613
Raja clavata	IZ	liver	21	(440 - 826)	(431.4 – 3820.1)	(0.000 - 2.885)	(13.88 - 81.629)	(0.001 - 1.255)	(0.000 - 0.090)	(0.000 - 0.029)	(0.261 - 2.476)
Daia davata	57	1	-	749 ± 68	2293.3 ± 650.1	1.539 ± 1.146	30.499 ± 4.233	0.346 ± 0.230	0.078 ± 0.025	0.006 ± 0.013	0.669 ± 0.521
Raja clavata	KZ	liver	5	(669 - 849)	(1567.7 – 3181.4)	(0.463 - 3.349)	(26.315 - 36.691)	(0.130 - 0.653)	(0.046 - 0.104)	(0.000 - 0.028)	(0.237 - 1.425)
Scyliorhinus	17	1	60	417 ± 58	244.1 ± 91.2	1.873 ± 1.727	29.800 ± 16.703	0.202 ± 0.220	0.059 ± 0.042	0.036 ± 0.064	1.129 ± 1.191
canicula	IZ	liver	60	(244 - 509)	(39.4 – 400.4)	(0.238 - 7.033)	(3.827 - 81.501)	(0.000 - 0.971)	(0.003 - 0.186)	(0.000 - 0.393)	(0.121 - 6.146)
Scyliorhinus	57	1	01	370 ± 62	182.7 ± 104.2	2.594 ± 2.362	27.772 ± 12.341	0.167 ± 0.149	0.068 ± 0.034	0.036 ± 0.037	0.496 ± 0.383
canicula	κz	liver	81	(244 - 500)	(41.3 – 485.4)	(0.466 - 14.108)	(2.900 - 63.125)	(0.001 - 0.857)	(0.026 - 0.168)	(0.000 - 0.169)	(0.077 - 2.182)

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
Congor congor	17	livor	20	696 ± 157	729.7 ± 525.3	0.934 ± 0.214	0.022 ± 0.045	0.069 ± 0.044	0.021 ± 0.043	0.096 ± 0.064
conger conger	IZ	liver	30	(416 - 1020)	(94.5 – 2161.8)	(0.280 - 1.361)	(0.000 - 0.186)	(0.000 - 0.169)	(0.000 - 0.151)	(0.003 - 0.331)
Congor congor	70	livor	21	691 ± 195	817.5 ± 672.6	1.119 ± 0.213	0.000 ± 0.000	0.102 ± 0.150	0.055 ± 0.079	0.131 ± 0.096
conger conger	κz	liver	21	(334 - 1010)	(50.8- 2341.7)	(0.768 - 1.553)	(0.000 - 0.000)	(0.008 - 0.657)	(0.000 - 0.208)	(0.006 - 0.368)
Merluccius	17	livor	40	364 ± 135	471.6 ± 534.3	1.69 ± 0.545	0.008 ± 0.027	0.015 ± 0.019	0.130 ± 0.229	0.034 ± 0.030
merluccius	IZ	liver	48	(233 - 795)	(97.7 - 2305.2)	(0.081 - 2.748)	(0.000 - 0.147)	(0.000 - 0.107)	(0.000 - 1.27)	(0.000 - 0.135)
Merluccius	70	livor	22	388 ± 51	469.7 ± 178.9	1.444 ± 0.383	0.014 ± 0.037	0.015 ± 0.025	0.022 ± 0.035	0.017 ± 0.012
merluccius	κz	liver	23	(284 - 476)	(147.3 – 858.4)	(0.924 - 2.629)	(0.000 - 0.113)	(0.000 - 0.117)	(0.000 - 0.095)	(0.002 - 0.042)
Daia alawata	17	livor	21	605 ± 116	1493.4 ± 940.0	0.798 ± 0.336	0.003 ± 0.014	0.015 ± 0.012	0.043 ± 0.062	0.051 ± 0.049
καjα ειανατα	IZ	liver	21	(440 - 826)	(431.4 – 3820.1)	(0.129 - 1.506)	(0.000 - 0.066)	(0.000 - 0.049)	(0.000 - 0.255)	(0.000 - 0.231)
Daia alawata	70	livor	-	749 ± 68	2293.3 ± 650.1	0.859 ± 0.185	0.013 ± 0.028	0.019 ± 0.008	0.036 ± 0.056	0.081 ± 0.044
καjα ειανατα	κz	liver	Э	(669 - 849)	(1567.7 – 3181.4)	(0.571 - 1.023)	(0.000 - 0.063)	(0.008 - 0.027)	(0.000 - 0.128)	(0.04 - 0.151)
Scyliorhinus	17	livor	60	417 ± 58	244.1 ± 91.2	0.873 ± 0.435	0.012 ± 0.042	0.026 ± 0.017	0.078 ± 0.084	0.038 ± 0.034
canicula	IZ	liver	60	(244 - 509)	(39.4 – 400.4)	(0.208 - 2.165)	(0.000 - 0.211)	(0.003 - 0.079)	(0.000 - 0.343)	(0.002 - 0.168)
Scyliorhinus	70	livor	01	370 ± 62	182.7 ± 104.2	1.147 ± 0.437	0.011 ± 0.026	0.029 ± 0.02	0.074 ± 0.084	0.129 ± 0.332
canicula	ĸΖ	nver	01	(244 - 500)	(41.3 – 485.4)	(0.488 - 3.143)	(0.000 - 0.112)	(0.007 - 0.151)	(0.000 - 0.442)	(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	Ν	Length (mm)	mass (g)	Total As	Asi	MMA	DMA	AsB
Distadus	17	2	264 ± 26	335 ± 108.2	6.23 ± 4.40 (2.34	0.033 ± 0.019	0.001 ± 0.001	0.004 ± 0.006	5.69 ± 4.39 (1.81 -
Diploaus sargus	IZ	3	(235 - 285)	(214.6 - 424)	- 11)	(0.013 - 0.048)	(0.000 - 0.002)	(0.000 - 0.011)	10.5)
Diplodus carque	70	7	245 ± 32	311.6 ± 121.5	11.73 ± 6.60 (2.8	0.037 ± 0.015	0.000 ± 0.000	0.001 ± 0.002	10.58 ± 6.71 (2.01 -
Dipiodus sargus	κz	/	(191 - 282)	(151 - 481.1)	- 22.5)	(0.022 - 0.059)	(0.000 - 0.000)	(0.000 - 0.005)	21.7)
Diplodus	17	F	227 ± 10	204.4 ± 33.6	4.44 ± 1.42 (2.64	0.06 ± 0.015	0.000 ± 0.000	0.005 ± 0.012	4.35 ± 1.56 (2.43 -
vulgaris	1Z	Э	(213 - 239)	(165.8 - 240)	- 6.41)	(0.037 - 0.073)	(0.000 - 0.000)	(0.000 - 0.026)	6.52)
Diplodus	50	0	213 ± 9	164.6 ± 19.3	8.93 ± 4.08 (4.42	0.081 ± 0.009	0.000 ± 0.001	0.007 ± 0.002	8.43 ± 4.23
vulgaris	κz	ō	(201 - 225)	(145.3 - 195.6)	- 15.9)	(0.069 - 0.099)	(0.000 - 0.002)	(0.002 - 0.009)	(3.80 - 15.5)
Helicolenus	17	F	186 ± 14	108.7 ± 24.6	5.52 ± 2.71 (2.46	0.021 ± 0.009	0.000 ± 0.000	0.001 ± 0.001	4.33 ± 2.45
dactylopterus	1Z	Э	(168 - 201)	(76.8 - 135)	- 9.18)	(0.014 - 0.033)	(0.000 - 0.000)	(0.000 - 0.002)	(1.79 - 7.61)
Helicolenus	57	c	191 ± 21	127 ± 36.3	11.38 ± 7.30 (3.7	0.04 ± 0.029	0.006 ± 0.01	0.001 ± 0.001	7.82 ± 5.74 (2.48 -
dactylopterus	RZ	0	(163 - 219)	(86.3 - 177.4)	- 23.6)	(0.017 - 0.095)	(0.000 - 0.019)	(0.000 - 0.002)	18.0)
Pagellus	17	7	269 ± 11	267.4 ± 38.8	6.03 ± 2.31 (2.99	0.057 ± 0.009	0.000 ± 0.001	0.006 ± 0.003	2.39 ± 0.71 (1.44 -
bogaraveo	1Z	/	(259 - 286)	(240.5 - 352.5)	- 9.9)	(0.044 - 0.072)	(0.000 - 0.002)	(0.000 - 0.01)	3.66)
Pagellus	50	7	240 ± 5	203.9 ± 12.4	5.67 ± 1.55 (3.51	0.049 ± 0.02	0.003 ± 0.007	0.009 ± 0.007	2.79 ± 1.31
bogaraveo	κz	/	(229 - 246)	(180.8 - 219.3)	- 8.31)	(0.029 - 0.09)	(0.000 - 0.018)	(0.000 - 0.022)	(2.02 - 5.67)
Pagellus	50	h	172 ± 3	68.3 ± 7.5	4.73 ± 1.51 (3.67	0.041 ± 0.001	0.006 ± 0.001	0.013 ± 0.007	4.58 ± 1.61 (3.44 -
erythrinus	ΠĽ	2	(170 - 174)	(63 - 73.6)	- 5.8)	(0.04 - 0.042)	(0.005 - 0.007)	(0.008 - 0.018)	5.71)
Paia clavata	17	E	604 ± 72	1474 ± 628.7	98.50 ± 19.94 (86	0.033 ± 0.015	0.053 ± 0.021	0.000 ± 0.000	95.7 ± 19.9 (80.1 -
καjα ειανατα	12	5	(497 - 699)	(677.6 - 2349.3)	- 132)	(0.016 - 0.057)	(0.021 - 0.072)	(0.000 - 0.000)	125)
Paia clavata	D7	r	744 ± 38	2293.3 ± 286	100.5 ± 27.58 (81	0.018 ± 0.003	0.084 ± 0.045	0.000 ± 0.000	110 ± 26.8 (91.4 -
καjα ειανατα	ΠĽ	2	(717 - 771)	(2091 - 2495.5)	- 120)	(0.016 - 0.021)	(0.052 - 0.116)	(0.000 - 0.000)	129)
Charus gurata	17	7	253 ± 10	233.9 ± 27.1	5.82 ± 3.29 (1.59	0.033 ± 0.026	0.000 ± 0.000	0.007 ± 0.003	5.91 ± 3.92 (0.971 -
Spurus uurutu	12	'	(242 - 272)	(204.9 - 289.9)	- 10.2)	(0.011 - 0.088)	(0.000 - 0.000)	(0.002 - 0.013)	11.6)
Sparus quireta	D7	r	261 ± 26	264.7 ± 99.5	12.62 ± 10.02	0.032 ± 0.002	0.000 ± 0.000	0.004 ± 0.005	13.1 ± 10.1 (5.99 -
spurus uurata	ĸΖ	Z	(243 - 280)	(194.4 - 335.1)	(5.53 - 19.7)	(0.031 - 0.033)	(0.000 - 0.000)	(0.000 - 0.007)	20.3)

Table 4: Pearson correlation coefficients between TE concentrations and length in the muscle and the liver of fish collected in this study. Significant699differences are highlighted by bold characters (*p < 0.05, **p < 0.01, ***p < 0.001)

700	7	0	0
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Species	Tissue	N IZ	N RZ	AI	As	Cd	Со	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

