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

Bouchoucha Marc [1, *], Chekri R. [2], Leufroy A. [2], Jitaru P. [2], Millour S. [2], Marchond N. [2], Chafey C. [2], Testu C. [2], Zinck J. [2], Cresson Pierre [3], Mirallès F. [1], Mahe A. [4], Arnich N. [4], Sanaa M. [4], Bemrah N. [4], Guérin T [2]

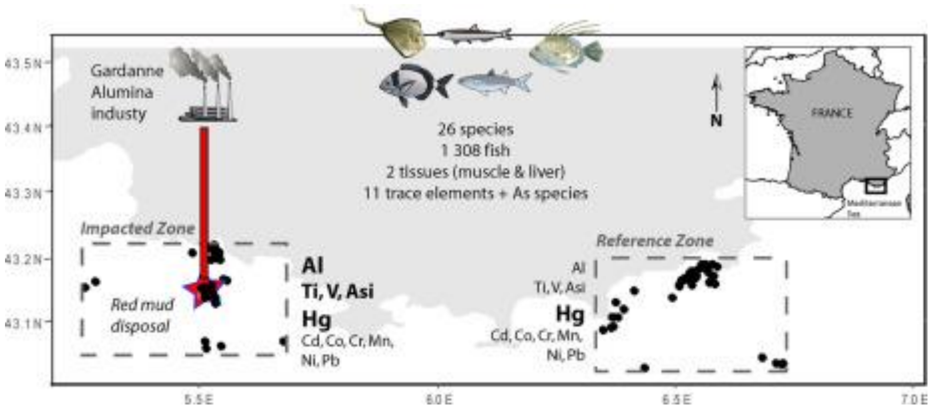
[1] Ifremer, Lab Environm Ressources Provence Azur Corse, CS 20330, F-83507 La Seyne Sur Mer, France
[2] Université Paris-Est, ANSES, Laboratory for Food Safety, F-94701 Maisons-Alfort, France
[3] Ifremer, Lab Ressources Halieutiques, BP 699, F-62 321 Boulogne sur Mer, France
[4] ANSES, Risk Assessment Directorate (DER), F-94701 Maisons-Alfort, France

* Corresponding author : Marc Bouchoucha, email address : marc.bouchoucha@ifremer.fr

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), aluminum (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 1308 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.

Graphical abstract



Highlights

► Human exposure to trace element contamination from industrial activity is of great concern. ► The Gardanne alumina refinery discharged ca. 20 Mt. of red mud into the Cassidaigne Canyon. ► 11 trace elements were analyzed in 1308 fish from the impacted zone and a reference zone. ► Red mud disposal in the Cassidaigne Canyon impacts fish contamination levels.

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 ‘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).

Fish sampling

Twenty-six commercial species were selected in this study with regard to their importance in the communities of both systems and in the diet of the local population. In addition, these species encompass a wide range of feeding habits and living environments. The sampling can thus be considered representative of all fish functional groups occurring in the area. The aim of the sampling program was to obtain similar species from both the impacted and the reference zone. Fish were 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 such as bottom and pelagic trawls, trammel nets, hooks, etc. The geographical coordinates of the 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 Co*nger 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.

Determination of trace element concentrations

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

Chemical speciation measurements were carried out on the seafood species in which total arsenic exposure was most prevalent: sea breams (gilthead sea bream *Sparus aurata*, black spot sea bream P*agellus bogaraveo*, common two-banded sea bream *Diplodus vulgaris*, white sea bream *Diplodus sargus* and red sea bream *Pagellus erythrinus*), redfish and rays. These species contribute more than 65% of total arsenic exposure for human consumers. For each species, between 2 and 8 composite samples per zone were analyzed. Each composite sample consisted of 3 to 5 sub-samples. Arsenic speciation was performed by HPLC/ICP-MS (HPLC Ultimate 300, Dionex, Voisins le bretonneux, France; ICP-MS X-Series^{II}, Thermo Scientifc, Courtaboeuf, France) coupling in standard mode after microwave-assisted extraction (Multiwave 3000, Anton-Paar). This method was validated in-house, accredited by COFRAC, and described by Leufroy et al. (2011). The following arsenic species were analyzed: As(III), As(V), monomethylarsonic acid (MA), dimethylarsinic acid (DMA), and arsenobetaine (AsB). For all these species, an LOD/LOQ of 0.002/0.005 mg As kg^{-1} wm was assessed under robust conditions. These speciation analyses were used to determine the levels of inorganic arsenic (sum of As(III) and As(V)). In parallel, the total arsenic concentration in the composite samples was also measured by the ICP-MS method described above for total TE analysis.

Several internal quality controls (IQCs) and associated criteria were used to ensure the reliability of the results. Each run included a standard calibration, blanks, several certified reference materials (CRMs): TORT-2 lobster hepatopancreas, ERM-CE278k mussel tissue and SRM 1548a typical diet, chosen to cover the 11 elements of interest (for total TE analysis) and BCR-627 tuna fish tissue (for As speciation analysis), two different spiked sample solutions, several fish samples including two 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 ($r^2 > 0.995$), blanks (values < limit of quantification (LOQ)), internal standards (values within 70 and 130% of the target value), mid-range standards (values within 80 and 120% of the target value), spiked standard solutions (spike recovery within 70 and 130% of the theoretical spiked standard value), CRMs (Z-score < ± 2) and duplicates (acceptable if the relative standard deviation (RSD) ≤ 20% when the mean value ≥ 5 x LOQ or RSD ≤ 40% when the mean value ≥ LOQ and < 5 x LOQ). When the acceptance criteria were not met, the 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.

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).

The only compound of inorganic As (Asi) found during this study was AS(III) while As(V) concentrations were systematically below LOD. Inorganic arsenic concentrations in fish muscle were not correlated with total As (AsT) concentrations (Table 3), and the average percentage of Asi (relative to AsT) ranged between 0.01 and 2.69% for all the species considered and in both sampling 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).*

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

Discussion

Very few studies have assessed the impact of the red mud disposal in the Cassidaigne Canyon on contamination levels in fish. To our knowledge, this is the first time that this has been investigated so thoroughly, with the analysis of 11 TE for a large number of individuals (n=1308) from 26 different fish species. In addition, two tissues (muscle and liver) were taken into account, comparing, when possible, contamination levels between the impacted zone and a reference one. Moreover, although several studies have already been conducted on TE in Mediterranean fish (e.g. Canli and Atli, 2003; Iamiceli et al., 2015; Mille et al., 2018), some elements such as Al, Ti and V have rarely been investigated (Eisler, 2010). Lastly, the present study provides a baseline dataset for various Mediterranean fish species and allows a preliminary investigation of certain factors affecting contamination.

A large number of the fish analyzed in this study presented concentration levels higher than the limits defined by the European Commission in fish muscle (EC, 2006). A total of 444 individuals from 15 different species (*C. conger, C. julis, D. sargus, D. vulgaris, G. melastomus, H. dactylopterus, M. merluccius, M. surmuletus, M. barbatus, P. bogaraveo, R. clavata, S. canicula, T. mediterraneus, 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 Hg in Mediterranean organisms (Cossa et al., 2012; Harmelin-Vivien et al., 2009; Koenig et al., 2013). The straightforward pattern observed for Hg in the present study may be largely driven by biological and biogeochemical mechanisms at play at the scale of the whole NE Mediterranean (Cossa and 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 0.05 mg.kg^{-1} wm for Pb and Cd, respectively), were lower, 8 and 19, respectively, which represent between 0.6 and 1.5% of the total number of fish analyzed. Between 75 and 90% of these threshold overruns concerned IZ, confirming the conclusions of Ourgaud et al. (2018), who found that these TE were more concentrated in *S. porcus* collected in the bay of Marseille (close to the Cassidaigne Canyon) than in those from the Gulf of Hyères. There is no regulatory limit for total As in fish muscle, but arsenic exposure is a significant worldwide environmental health concern. Overall, the data we 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, *C. julis*, *L. ramada* and *L. aurata* were captured between 5 and 35 m depth whereas *G. melastomus*, *H. dactylopterus* and *P. bogaraveo* were caught between 141 and 391m depth. This may explain a part of the inter-specific variations in TE contamination and the lower TE concentrations generally observed in *L. ramada*, *L. aurata* and *C. julis* (Table 1), Finally, inter-specific differences in TE concentrations may also be driven by contrasting life-history and behavior. Factors such as preferred habitat (*e.g*. benthic and pelagic), longevity, diet and foraging behavior (*e. g.* vertical migrations and type of prey consumed), metabolic rates and biochemical response (Burger, 2007; Zhao et al., 2012) have been demonstrated to affect rates of TE uptake and excretion from both food and water pathways. Previous works showed that species that are continuously in contact with contaminants in sediment have developed higher detoxication abilities, resulting in lower body burdens while being exposed to the same contaminant load as pelagic species (Sole et al., 2006). This may explain why TE concentrations in the benthic ray *R. clavata* are genarally lower than those o the demersal *S. canicula* (Table1, 2), despite similar trophic levels (Froese and Pauly, 2014; Stergiou and Karpouzi, 2001) and living depth (both species were captured between 100 and 380m depth).

Fish length is often recognized to be of importance in determining the rate of physiological processes that influence uptake, distribution and elimination of TE (Canli and Atli, 2003). However, except for Hg, our analysis revealed no conclusive evidence that mean TE concentrations were systematically linked to fish length, which is in good agreement with the results reported in the literature. Indeed, although it is generally accepted that Hg load in fish body is strongly affected by size it is still not clear if the other TE accumulations are size-dependent. This is supported by the fact that most TE (with the exception of As and Hg) do not biomagnify along aquatic food webs (Fey et al., 2019; Xu and Wang, 2002). The available literature on the correlation between TE concentrations in muscle tissue and fish length and weight have shown that a dependency exists for some species (e.g. Gobert et al., 2017), while for other species, no dependency is found (e.g. Mille et al., 2018). The absence of relationship likely indicates that fish regulate some TE at a certain concentration required for efficient metabolic activities (Canli and Atli, 2003; Hornung et al., 1993). Moreover, when significant, the relationships between TE (other than Hg) and fish lengths are generally negative (Endo et al., 2008). This may result from a higher metabolic activity in fast-growing young individuals, requiring higher amounts of these elements than older individuals or through a higher excretion rate and/or dilution of metal burden with growth, and lower metabolism (Eisler, 2010). In addition, length is commonly used as a proxy of age and chronic exposure to contaminants, as length is quickly measured, while accurate age determination requires complex and time-consuming procedures (*e.g.* otolith or vertebrae reading for teleost and chondrichtyans, respectively). This pattern is nonetheless blurred by the fact that growth rate affects the age - length relationship. As a result, individuals of the same length but from zones with different nutritional inputs (and thus different growth rates) may have different age and may have been thus exposed to contaminant during different periods of time.

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, 2001; Uluturhan and Kucuksezgin, 2007). With the exception of Hg, As and Ti, the liver to muscle concentration ratios calculated in this study were generally over 1 and reached up to 360. This is not surprising because, in general, TE are preferentially accumulated in the liver of aquatic organisms (Henry et al., 2004), which likely results from detoxification processes (Eisler, 2010). For Hg, higher concentrations are commonly found in fish muscle. Indeed, methylmercury is the predominant form of Hg in fish, ranging from 80% to 100% (Bloom, 1992; Chouvelon et al., 2018; Magalhaes et al., 2007) and is mainly accumulated in fish muscle due to its affinity to protein (Amlund et al., 2007; Harris et al., 2003). This difference in chemical affinity explains why the liver to muscle concentration ratios for Hg ranged here from 0.47 to 0.97. Considering As, Eisler (2010) stated that hepatic concentrations are usually higher than those found in muscle tissues, a pattern not observed in the present study as the liver to muscle concentration ratios ranged between 0.34 and 0.90. Such ratios below 1 have already been reported in benthic (*Pteromylaeus bovinus* and *Myliobatis aquila*) and pelagic (*Pteroplatytrygon violacea*) rays from the northern Adriatic Sea (Slejkovec et al., 2014), in *S.*

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

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

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References

Amiard JC, Amiardtriquet C, Berthet B, Metayer C. Comparative study of the patterns of bioaccumulation of essential (Cu, Zn) and non-essential (Cd, Pb) trace metals in various estuarine and coastal organisms. Journal of Experimental Marine Biology and Ecology 1987; 106: 73-89.

Amlund H, Lundebye AK, Berntssen MHG. Accumulation and elimination of methylmercury in Atlantic cod (*Gadus morhua* L.) following dietary exposure. Aquatic Toxicology 2007; 83: 323-330.

Anderson JM. Permutation tests for univariate or multivariate analysis of variance and regression. Canadian Journal of Fisheries and Aquatic Sciences 2001; 58: 626-639.

ANSES. Note d'appui scientifique et technique de l'Agence nationale de sécurité sanitaire de l'alimentation , de l'environnement et du travail relatif à l'impact potentiel sur la santé humaine du rejet en Méditerranée d'effluents issus des activités de transformation de minerai de bauxite, 2016, pp. 74. https://www.anses.fr/en/system/files/ERCA2014SA0223.pdf

Asante KA, Agusa T, Kubota R, Mochizuki H, Ramu K, Nishida S, et al. Trace elements and stable isotope ratios (delta C-13 and delta N-15) in fish from deep-waters of the Sulu Sea and the Celebes Sea. Marine Pollution Bulletin 2010; 60: 1560-1570.

Benjamini Y, Hochberg Y. Controlling the false discovery rate: a practical and powerful approach to multiple testing. Journal of the Royal statistical society: series B (Methodological) 1995; 57: 289-300.

Bloom NS. On the chemical form of mercury in edible fish and marine invertebrate tissue. Canadian Journal of Fisheries and Aquatic Sciences 1992; 49: 1010-1017.

Bouchoucha M, Brach-Papa C, Gonzalez J-L, Lenfant P, Darnaude A. Growth, condition and metal concentration in juveniles of two *Diplodus* species in ports. Marine Pollution Bulletin 2018; 126: 31-42.

Bourcier M. Ecoulement des boues rouges dans le canyon de la Cassidaigne (décembre 1968). TeHthys, France 1969; 1: 779.

Bourcier M, Zibrowius H. Les «boues rouges» déversées dans le canyon de la Cassidaigne (région de Marseille). Observations en soucoupe plongeante SP 1973; 350: 811-842.

Bryan G. The effects of heavy metals (other than mercury) on marine and estuarine organisms. Proceedings of the Royal Society of London B: Biological Sciences 1971; 177: 389-410.

Bryan G, Waldichuk M, Pentreath R, Darracott A. Bioaccumulation of marine pollutants. Philosophical Transactions of the Royal Society of London. Series B, Biological Sciences 1979: 483-505.

Burger J. A framework and methods for incorporating gender-related issues in wildlife risk assessment: Gender-related differences in metal levels and other contaminants as a case study. Environmental Research 2007; 104: 153-162.

Canli M, Atli G. The relationships between heavy metal (Cd, Cr, Cu, Fe, Pb, Zn) levels and the size of six Mediterranean fish species. Environmental Pollution 2003; 121: 129-136.

Castro-Gonzalez MI, Mendez-Armenta M. Heavy metals: Implications associated to fish consumption. Environmental Toxicology and Pharmacology 2008; 26: 263-271.

Chapman DV. Water quality assessments: a guide to the use of biota, sediments, and water in environmental monitoring: E & Fn Spon London, 1996.

Chevallier E, Chekri R, Zinck J, Guérin T, Noël L. Simultaneous determination of 31 elements in foodstuffs by ICP-MS after closed-vessel microwave digestion: Method validation based on the accuracy profile. Journal of Food Composition and Analysis 2015; 41: 35-41.

Chouvelon T, Cresson P, Bouchoucha M, Brach-Papa C, Bustamante P, Crochet S, et al. Oligotrophy as a major driver of mercury bioaccumulation in medium-to high-trophic level consumers: A marine ecosystem-comparative study. Environmental Pollution 2018; 233: 844-854.

Chouvelon T, Spitz J, Caurant F, Mendez-Fernandez P, Autier J, Lassus-Debat A, et al. Enhanced bioaccumulation of mercury in deep-sea fauna from the Bay of Biscay (north-east Atlantic) in relation to trophic positions identified by analysis of carbon and nitrogen stable isotopes. Deep-Sea Research Part I-Oceanographic Research Papers 2012; 65: 113-124.

Clarke KR, Warwick RM. Change in marine communities: an approach to statistical analysis and interpretation. PRIMER-E Limited Plymouth, UK, 2001.

Copat C, Arena G, Fiore M, Ledda C, Fallico R, Sciacca S, et al. Heavy metals concentrations in fish and shellfish from eastern Mediterranean Sea: Consumption advisories. Food and Chemical Toxicology 2013; 53: 33-37.

Cossa D, Coquery M. The Mediterranean mercury anomaly, a geochemical or a biological issue. In: Saliot A, editor. The Mediterranean Sea. Springer Berlin Heidelberg, Berlin, Heidelberg, 2005, pp. 177-208.

Cossa D, Harmelin-Vivien M, Mellon-Duval C, Loizeau V, Averty B, Crochet S, et al. Influences of bioavailability, trophic position, and growth on methylmercury in hakes (*Merluccius merluccius*) from northwestern Mediterranean and northeastern Atlantic. Environmental Science & Technology 2012; 46: 4885-4893.

Cresson P, Bouchoucha M, Miralles F, Elleboode R, Mahe K, Marusczak N, et al. Are red mullet efficient as bio-indicators of mercury contamination? A case study from the French Mediterranean. Marine Pollution Bulletin 2015; 91: 191-199.

Cresson P, Fabri MC, Bouchoucha M, Papa CB, Chavanon F, Jadaud A, et al. Mercury in organisms from the Northwestern Mediterranean slope: Importance of food sources. Science of The Total Environment 2014a; 497: 229-238.

Cresson P, Ruitton S, Ourgaud M, Harmelin-Vivien M. Contrasting perception of fish trophic level from stomach content and stable isotope analyses: A Mediterranean artificial reef experience. Journal of Experimental Marine Biology and Ecology 2014b; 452: 54-62.

Damiano S, Papetti P, Menesatti P. Accumulation of heavy metals to assess the health status of swordfish in a comparative analysis of Mediterranean and Atlantic areas. Marine Pollution Bulletin 2011; 62: 1920-1925.

Dauvin JC. Towards an impact assessment of bauxite red mud waste on the knowledge of the structure and functions of bathyal ecosystems: The example of the Cassidaigne canyon (north-western Mediterranean Sea). Marine Pollution Bulletin 2010; 60: 197-206.

De Gieter M, Leermakers M, Van Ryssen R, Noyen J, Goeyens L, Baeyens W. Total and toxic arsenic levels in North Sea fish. Archives of Environmental Contamination and Toxicology 2002; 43: 406-417.

Denton GRW, Concepcion LP, Wood HR, Morrison RJ. Trace metals in marine organisms from four harbours in Guam. Marine Pollution Bulletin 2006; 52: 1784-1804.

Dural M, Goksu MZL, Ozak AA, Derici B. Bioaccumulation of some heavy metals in different tissues of *Dicentrarchus labrax* L, 1758, *Sparus aurata* L, 1758 and *Mugil cephalus* L, 1758 from the Camlik lagoon of the eastern cost of Mediterranean (Turkey). Environmental Monitoring and Assessment 2006; 118: 65-74.

EC. Commission Regulation (EC) No 1881/2006 of 19 December 2006 setting maximum levels for certain contaminants in foodstuffs. Off J Eur Union 2006; 364: 5-24.

Eisler R. Chapter 3 - Fishes. In: Eisler R, editor. Compendium of Trace Metals and Marine Biota. Elsevier, Amsterdam, 2010, pp. 39-220.

Ellis DV, Poling GW, Baer RL. Submarine tailings disposal (STD) for mines: An introduction. Marine Georesources & Geotechnology 1995; 13: 3-18.

Endo T, Hisamichi Y, Haraguchi K, Kato Y, Ohta C, Koga N. Hg, Zn and Cu levels in the muscle and liver of tiger sharks (*Galeocerdo cuvier*) from the coast of Ishigaki Island, Japan: Relationship between metal concentrations and body length. Marine Pollution Bulletin 2008; 56: 1774-1780.

Fabri MC, Pedel L, Beuck L, Galgani F, Hebbeln D, Freiwald A. Megafauna of vulnerable marine ecosystems in French mediterranean submarine canyons: Spatial distribution and anthropogenic impacts. Deep-Sea Research Part Ii-Topical Studies in Oceanography 2014; 104: 184-207.

Fattorini D, Alonso-Hernandez CM, Diaz-Asencio M, Munoz-Caravaca A, Pannacciulli FG, Tangherlini M, et al. Chemical special-lion of arsenic in different marine organisms: Importance in monitoring studies. Marine Environmental Research 2004; 58: 845-850.

Fernandes C, Fontainhas-Fernandes A, Peixoto F, Salgado MA. Bioaccumulation of heavy metals in *Liza saliens* from the Esmoriz-Paramos coastal lagoon, Portugal. Ecotoxicology and Environmental Safety 2007; 66: 426-431.

Fey P, Bustamante P, Bosserelle P, Espiau B, Malau A, Mercader M, et al. Does trophic level drive organic and metallic contamination in coral reef organisms? Science of the Total Environment 2019; 667: 208-221.

Fontanier C, Fabri MC, Buscail R, Biscara L, Koho K, Reichart GJ, et al. Deep-sea foraminifera from the Cassidaigne Canyon (NW Mediterranean): Assessing the environmental impact of bauxite red mud disposal. Marine Pollution Bulletin 2012; 64: 1895-1910.

Francesconi KA. Arsenic species in seafood: Origin and human health implications. Pure and Applied Chemistry 2010; 82: 373-381.

Froese R, Pauly D. Fishbase. Available: www.fishbase.org, 2014.

Gatti J, Petrenko A, Devenon JL, Leredde Y, Ulses C. The Rhone river dilution zone present in the northeastern shelf of the Gulf of Lion in December 2003. Continental Shelf Research 2006; 26: 1794-1805.

Gobert S, Pasqualini V, Dijoux J, Lejeune P, Durieux EDH, Marengo M. Trace element concentrations in the apex predator swordfish (*Xiphias gladius*) from a Mediterranean fishery and risk assessment for consumers. Marine Pollution Bulletin 2017; 120: 364-369.

Gray JE, Greaves IA, Bustos DM, Krabbenhoft DP. Mercury and methylmercury contents in mine-waste calcine, water, and sediment collected from the Palawan Quicksilver Mine, Philippines. Environmental Geology 2003; 43: 298-307.

Harmelin-Vivien M, Cossa D, Crochet S, Banaru D, Letourneur Y, Mellon-Duval C. Difference of mercury bioaccumulation in red mullets from the north-western Mediterranean and Black seas. Marine Pollution Bulletin 2009; 58: 679-685.

Harris HH, Pickering IJ, George GN. The chemical form of mercury in fish. Science 2003; 301: 1203-1203.

Hellou J, Zitko V, Friel J, Alkanani T. Distribution of elements in tissues of yellowtail flounder *Pleuronectes ferruginea*. Science of The Total Environment 1996; 181: 137-146.

Henry F, Amara R, Courcot L, Lacouture D, Bertho ML. Heavy metals in four fish species from the French coast of the eastern English Channel and Southern Bight of the North Sea. Environment International 2004; 30: 675-683.

Hindmarsh JT. Arsenic, its clinical and environmental significance. The Journal of Trace Elements in Experimental Medicine: The Official Publication of the International Society for Trace Element Research in Humans 2000; 13: 165-172.

Hornung H, Krom MD, Cohen Y, Bernhard M. Trace-metam content in deep-water sharks from the eastern Mediterranean-Sea. Marine Biology 1993; 115: 331-338.

Iamiceli A, Ubaldi A, Lucchetti D, Brambilla G, Abate V, De Felip E, et al. Metals in Mediterranean aquatic species. Marine Pollution Bulletin 2015; 94: 278-283.

Koenig S, Solé M, Fernández-Gómez C, Díez S. New insights into mercury bioaccumulation in deep-sea organisms from the NW Mediterranean and their human health implications. Science of The Total Environment 2013; 442: 329-335.

Leufroy A, Noël L, Dufailly V, Beauchemin D, Guérin T. Determination of seven arsenic species in seafood by ion exchange chromatography coupled to inductively coupled plasma-mass spectrometry following microwave assisted extraction: method validation and occurrence data. Talanta 2011; 83: 770-779.

Long AM, Wang WX. Metallothionein induction and bioaccumulation kinetics of Cd and Ag in the marine fish Terapon jarbua challenged with dietary or waterborne Ag and Cu. Marine Ecology Progress Series 2005; 291: 215-226.

Magalhaes MC, Costa V, Menezes GM, Pinho MR, Santos RS, Monteiro LR. Intra- and inter-specific variability in total and methylmercury bioaccumulation by eight marine fish species from the Azores. Marine Pollution Bulletin 2007; 54: 1654-1662.

Malea P, Haritonidis S. Concentration of Aluminium in *Halophila stipulacea* (Forsk.) archers. and the substrate of the Antikyra Gulf, Greece. Toxicological & Environmental Chemistry 1989; 20-21: 241-248.

Mason RP. Trace metals in aquatic systems: John Wiley & Sons, 2013.

Metian M, Warnau M, Chouvelon T, Pedraza F, Baena A, Bustamante P. Trace element bioaccumulation in reef fish from New Caledonia: Influence of trophic groups and risk assessment for consumers. Marine Environmental Research 2013; 87-88: 26-36.

Mille T, Cresson P, Chouvelon T, Bustamante P, Brach-Papa C, Bruzac S, et al. Trace metal concentrations in the muscle of seven marine species: Comparison between the Gulf of Lions (North-West Mediterranean Sea) and the Bay of Biscay (North-East Atlantic Ocean). Marine Pollution Bulletin 2018; 135: 9-16.

Millero FJ, Woosley R, Ditrolio B, Waters J. Effect of Ocean Acidification on the Speciation of Metals in Seawater. Oceanography 2009; 22: 72-85.

Millour S, Noël L, Chekri R, Vastel C, Kadar A, Guérin T. Internal quality controls applied in inductively coupled plasma mass spectrometry multi-elemental analysis in the second French Total Diet Study. Accreditation and quality assurance 2010; 15: 503-513.

Miniero R, Abate V, Brambilla G, Davoli E, De Felip E, De Filippis SP, et al. Persistent toxic substances in Mediterranean aquatic species. Science of The Total Environment 2014; 494: 18-27.

Morford JL, Emerson S. The geochemistry of redox sensitive trace metals in sediments. Geochimica Et Cosmochimica Acta 1999; 63: 1735-1750.

Mormede S, Davies IM. Heavy metal concentrations in commercial deep-sea fish from the Rockall Trough. Continental Shelf Research 2001; 21: 899-916.

Neff JM. Ecotoxicology of arsenic in the marine environment. Environmental Toxicology and Chemistry 1997; 16: 917-927.

Nriagu JO, Pacyna JM. Quantitative assessment of worldwide contamination of air, water and soils by trace-metals. Nature 1988; 333: 134-139.

Ourgaud M, Ruitton S, Bourgogne H, Bustamante P, Churlaud C, Guillou G, et al. Trace elements in a Mediterranean scorpaenid fish: Bioaccumulation processes and spatial variations. Progress in Oceanography 2018; 163: 184-195.

Pagano G, Meric S, De Biase A, Iaccarino M, Petruzzelli D, Tunay O, et al. Toxicity of bauxite manufacturing by-products in sea urchin embryos. Ecotoxicology and Environmental Safety 2002; 51: 28-34.

Percin F, Sogut O, Altinelataman C, Soylak M. Some trace elements in front and rear dorsal ordinary muscles of wild and farmed bluefin tuna (Thunnus thynnus L. 1758) in the Turkish part of the eastern Mediterranean Sea. Food and Chemical Toxicology 2011; 49: 1006-1010.

R Core Team. R: A Language and Environment for Statistical Computing. R Foundation for Statistical Computing, Vienna, Austria, 2014.

Rahman MA, Hasegawa H, Lim RP. Bioaccumulation, biotransformation and trophic transfer of arsenic in the aquatic food chain. Environmental Research 2012; 116: 118-135.

Raimundo J, Anes B, Caetano M, Giacomello E, Menezes G, Vale C. Trace-element concentrations in muscle and liver of 11 commercial fish species from Condor Seamount, Azores Archipelago (Portugal). Deep-Sea Res 2013; 98: 137-147.

Raimundo J, Vale C, Martins I, Fontes J, Graca G, Caetano M. Elemental composition of two ecologically contrasting seamount fishes, the bluemouth (*Helicolenus dactylopterus*) and blackspot seabream (*Pagellus bogaraveo*). Marine Pollution Bulletin 2015; 100: 112-121.

Saha JC, Dikshit AK, Bandyopadhyay M, Saha KC. A review of arsenic poisoning and its effects on human health. Critical Reviews in Environmental Science and Technology 1999; 29: 281-313.

Slejkovec Z, Stajnko A, Falnoga I, Lipej L, Mazej D, Horvat M, et al. Bioaccumulation of Arsenic Species in Rays from the Northern Adriatic Sea. International Journal of Molecular Sciences 2014; 15: 22073-22091.

Sole M, de la Parra LMG, Alejandre-Grimaldo S, Sarda F. Esterase activities and lipid peroxidation levels in offshore commercial species of the NW Mediterranean Sea. Marine Pollution Bulletin 2006; 52: 1708-1716.

Squadrone S, Burioli E, Monaco G, Koya MK, Prearo M, Gennero S, et al. Human exposure to metals due to consumption of fish from an artificial lake basin close to an active mining area in Katanga (DR Congo). Science of The Total Environment 2016; 568: 679-684.

Stergiou KI, Karpouzi VS. Feeding habits and trophic levels of Mediterranean fish. Reviews in Fish Biology and Fisheries 2001; 11: 217-254.

Storelli MM, Busco VP, Marcotrigiano GO. Mercury and arsenic speciation in the muscle tissue of *Scyliorhinus canicula* from the Mediterranean sea. Bulletin of Environmental Contamination and Toxicology 2005; 75: 81-88.

Storelli MM, Marcotrigiano GO. Organic and inorganic arsenic and lead in fish from the South Adriatic Sea, Italy. Food Additives and Contaminants 2000; 17: 763-768.

Suarez-Serrano A, Alcaraz C, Ibanez C, Trobajo R, Barata C. *Procambarus clarkii* as a bioindicator of heavy metal pollution sources in the lower Ebro River and Delta. Ecotoxicology and Environmental Safety 2010; 73: 280-286.

Swales S, Storey AW, Roderick ID, Figa BS, Bakowa KA, Tenakanai CD. Biological monitoring of the impacts of the Ok Tedi copper mine on fish populations in the Fly River system, Papua New Guinea. Science of The Total Environment 1998; 214: 99-111.

Turkmen M, Turkmen A, Tepe Y, Tore Y, Ates A. Determination of metals in fish species from Aegean and Mediterranean seas. Food Chemistry 2009; 113: 233-237.

Uluturhan E, Kucuksezgin F. Heavy metal contaminants in Red Pandora (*Pagellus erythrinus*) tissues from the Eastern Aegean Sea, Turkey. water research 2007; 41: 1185-1192.

Vallee BL, Auld DS. Zinc coordination, function, and structure of zinc enzymes and other proteins. Biochemistry 1990; 29: 5647-5659.

Williams TM, Weeks JM, Apostol AN, Miranda CR. Assessment of mercury contamination and human exposure associated with coastal disposal of waste from a cinnabar mining operation, Palawan, Philippines. Environmental Geology 1999; 39: 51-60.

Xu Y, Wang WX. Exposure and potential food chain transfer factor of Cd, Se and Zn in marine fish *Lutjanus argentimaculatus*. Marine Ecology Progress Series 2002; 238: 173-186.

Zhao S, Feng CH, Quan WM, Chen XF, Niu JF, Shen ZY. Role of living environments in the accumulation characteristics of heavy metals in fishes and crabs in the Yangtze River Estuary, China. Marine Pollution Bulletin 2012; 64: 1163-1171.

Figures

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

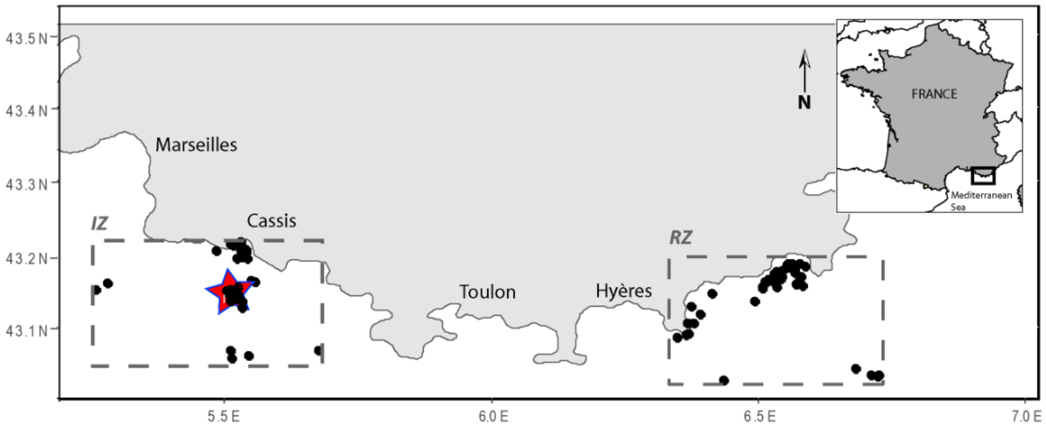


Table 1: Mean TE concentrations ± standard deviation (in mg.kg^{-1} wm) measured in fish muscle from the red mud impacted (IZ) and the reference (RZ) zones. Minimum and maximum values are given in brackets.

Species	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)

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)

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

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 (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)

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)

Zeus faber	RZ	muscle	1	355	612.2	0.059	0.000	0.004	0.000	0.000

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	**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)

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)

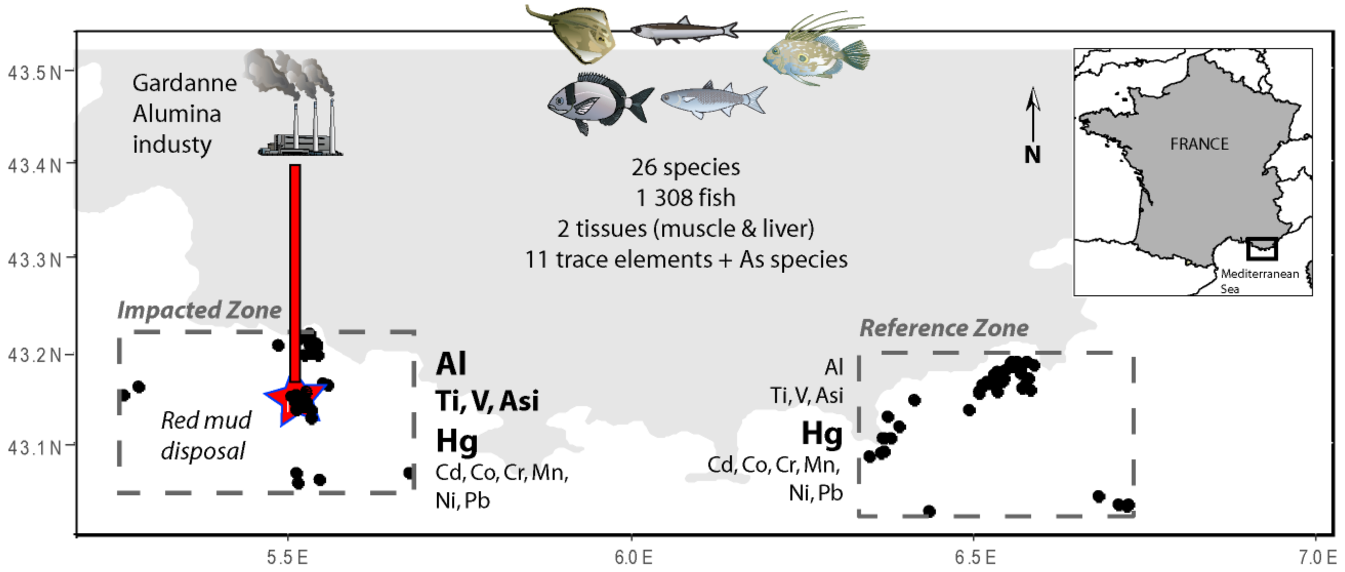
Table 3: Speciation of arsenic in muscle of fish collected from the zone (IZ) impacted by red mud and the reference zone (RZ) (in mg.kg^{-1} wm). Asi = inorganic arsenic (= As(III) because As(V) was not found in this study), MMA = monomethylarsonic acid, DMA = dimethylarsinic acid, AsB = arsenobetaine.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



Highlights

- Human exposure to trace element contamination from industrial activity is of great concern.
- The Gardanne alumina refinery discharged *ca*. 20 Mt of red mud into the Cassidaigne Canyon.
- 11 trace elements were analyzed in 1308 fish from the impacted zone and a reference zone.
- Red mud disposal in the Cassidaigne Canyon impacts fish contamination levels.