

# First assessment of Rare Earth Element organotropism in *Solea solea* in a coastal area: The West Gironde Mud Patch (France)

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## Abstract

Few studies exist on bioaccumulation and internal distribution of Rare Earth Elements (REEs) in marine fishes. REEs organotropism was determined in common sole (*Solea solea*) from the West Gironde Mud Patch (WGMP; N-E Atlantic Coast, France). The highest REEs concentrations occurred in liver ( $213 \pm 49.8 \mu\text{g kg}^{-1} \text{DW}$ ) and gills ( $119 \pm 77.5 \mu\text{g kg}^{-1} \text{DW}$ ) followed by kidneys ( $57.7 \pm 25.5 \mu\text{g kg}^{-1} \text{DW}$ ), whereas the lowest levels were in muscles ( $4.38 \pm 1.20 \mu\text{g kg}^{-1} \text{DW}$ ) of *Solea solea*. No significant age- or sex-related differences were observed. The organotropism varied among groups of REEs. Light and heavy REEs preferentially accumulated in liver and gills, respectively. All considered organs showed different normalized REEs patterns, suggesting differences in internal distribution processes between organs. Further work should address: (1) baseline levels worldwide, and (2) factors controlling uptake and organ-specific bioaccumulation of REEs.

## Keywords

REEs, bioaccumulation, organ-specific signature, Fish, RIOMAR

# 1. Introduction

Through their sedimentary and biogeochemical characteristics, River-dominated Ocean MARGin (RIOMAR) systems constitute key components of ecosystem functioning in coastal areas. They are especially important for the delivery of ecosystem services to mankind (Charuau, 1988; Lamarque et al., 2021). The West Gironde Mud Patch (WGMP) is a sedimentary body located in the Bay of Biscay (France), which is supplied in particles by Gironde Estuary inputs. The WGMP is a key area for early life stages of several exploited marine species, including the common sole (*Solea solea*), an emblematic species representing a strong economic interest for fisheries.

Many coastal species, including algae, bivalves, crustaceans, fishes, and mammals are increasingly exposed to emerging metal contaminants, such as the Rare Earth Elements (REEs; MacMillan et al., 2017; Squadrone et al., 2019). The current knowledge regarding health and environmental risks associated with the exposure to these emerging contaminants remains however largely insufficient (Neira et al., 2022; Piarulli et al., 2021).

REEs represent a group of 17 trace metals, including scandium (Sc;  $Z = 21$ ), yttrium (Y;  $Z = 39$ ) and the 15 lanthanides from lanthanum (La;  $Z = 57$ ) to lutetium (Lu;  $Z = 71$ ). REEs are lithophilic metals with similar physical and chemical properties, making them a homogeneous group in the environment due to their close electronic configurations and stable +3 oxidation state (Long et al., 2012). They are usually divided into light REEs (LREEs; from La to Nd), medium REEs (MREEs; from Sm to Gd) and heavy REEs (HREEs; from Tb to Lu). The electronic, magnetic, optical, or catalytic properties of REEs are widely used in high-technology, chemical industry, and medical applications (Bru et al., 2015). The recent and rapid expansion in REEs consumption in a wide variety of modern applications (e.g. medical technology, green energy, automobile) has led to an increase in their release into riverine (e.g. Kulaksız and Bau, 2013; Li et al., 2015; Johannesson et al., 2017; Lerat-Hardy et al., 2019) and

coastal/marine (e.g. Pereto et al., 2023; Brito et al., 2018; Tranchida et al., 2011) environments, making them emerging contaminants.

There is evidence that the Gironde Estuary receives increasing inputs of anthropogenic Gd, from urban areas mainly *via* the Garonne River, reflecting the evolution of national anti-cancer strategies (Lerat-Hardy et al., 2019). Gadolinium anomalies have been measured in freshwater clams (*Corbicula fluminea*) in a urban tributary of the Gironde Estuary (Pereto et al., 2020). In addition, Briant et al. (2021) have reported the highest REEs concentrations in the wild oysters of the Gironde Estuary mouth compared to those collected along the French Atlantic and Mediterranean coasts. Conversely, no data are currently available on potential REEs releases into the coastal zone of the Gironde Estuary. Hence, there is a real need for monitoring natural REEs levels in coastal organisms living in this zone, such as the common sole.

Although some recent studies have addressed the natural bioaccumulation of REEs in marine fishes (Li et al., 2016; Squadrone et al., 2019; Squadrone et al., 2020; Lortholarie et al., 2020), only few studies have investigated REEs distribution in different tissues (organotropism), and the age or sex effects on their bioaccumulation (Squadrone et al., 2020; Lortholarie et al., 2021; Squadrone et al., 2022). This constitutes a major gap, since organotropism provides key information for (i) the identification of transfer pathways, (ii) the understanding of detoxification at the individual scale, and (iii) the evaluation of potential health risks related to the consumption of specific tissues. Moreover, to our knowledge, there is no data available for REEs in *Solea solea*.

The objectives of this study are to i) evaluate the potential influence of sex and age on REEs bioaccumulation, ii) provide the first data on the natural REEs concentrations in the muscles, kidneys, liver and gills of the common sole, *Solea solea*, and compare with previous studies on fish, iii) assess the contribution of REEs between these target organs, and iv) compare REEs patterns between target organs.

## 2. Materials and methods

### 2.1. Study area

The West Gironde Mud Patch (WGMP) is a ca 420 km<sup>2</sup> large thin sedimentary body about 15 km off the Gironde Estuary within the Bay of Biscay (France). The WGMP extends over depth ranging from 30 to 75 m along a main NE-SW orientation (Figure 1; Jouanneau et al., 1989; Lesueur et al., 1991). The WGMP is a temperate coastal system supplied in particles by the Gironde Estuary (Dubosq et al., 2021; Lamarque et al., 2021) and strongly submitted to hydrodynamic influences (Lesueur et al., 2002). Sandy-muddy environments, such as the WGMP, are habitats for several coastal species, including the common sole *Solea solea* (Koutsikopoulos et al., 1989).

### 2.2. Sampling

*Solea solea* were collected in April 2021 in the distal area of WGMP during the MAGMA cruise (MAGMA; 10.17600/18000861) which took place on board the OR/V “Côtes de la Manche”. A total of 14 individuals were collected using a beam trawl and kept alive on board within trays fueled with running seawater until the return to the harbor. They were immediately euthanized, visually sexed, weighed (wet weight), measured (total length) and dissected. Target organs (i.e. liver, kidneys, gills, and muscles) were recovered, weighed and kept at -20°C before freeze-drying them in the laboratory. The otoliths, located in the inner ear cavity, were collected to determine the age of individuals, following the international aging protocol (Vitale et al., 2019).

### 2.3. Analytical methods

All laboratory material in contact with the samples was cleaned by immersion into HNO<sub>3</sub> (10% v/v) for three days, thoroughly rinsed with ultrapure water (MilliQ®), dried in a laminar flow hood and stored in double sealed polypropylene bags until use. Freeze-dried organs were ground and homogenized in an agate mortar. Subsamples of each organ (0.100-0.150 g DW) and certified reference material (CRM) were microwave digested (MARS 6, CEM) in Teflon

tubes with 4 mL/4 mL of HNO<sub>3</sub> ultrex/ MilliQ® (Nørregaard et al., 2019). Samples, blanks, and CRM were diluted using HNO<sub>3</sub> 2% before analysis. The simultaneous determination of the REEs concentrations was performed by a Triple Quadrupole Inductively Coupled Plasma-Mass Spectrometer (Thermo Scientific® iCAP TQ - ICP-MS) in KED and O<sub>2</sub> modes, depending on the isotope. External calibration was used with multi-element mixed standard solutions, prepared for each analytical session. Interference control and correction were performed according to Lerat-Hardy et al. (2019). After blank correction, REEs concentrations were expressed in microgram per kilogram dry weight of tissue samples ( $\mu\text{g kg}^{-1}$  DW).

#### 2.4. Analytical quality control

The systematic parallel analysis of procedural blanks and the CRM (SLRS-6; Yeghicheyan et al., 2019, BCR-668 muscles of mussels; Kramer et al., 2001) allowed for the determination of detection limits, accuracy, and precision. Results obtained were in good agreement with the certified values. Recovery values for BCR-668 ranged from 76 to 93% and between 84 and 109 % for SLRS-6. All the relative standard deviations were below 10% for all REEs. Limits of detection (LOD; 3  $\sigma$  of procedural blanks) were determined for each REEs (supplementary table 1).

#### 2.5. Data processing and statistical analysis

##### *2.5.1. Data treatment for comparison with other studies*

REEs concentrations in target organs of *Solea solea* from the WGMP have been compared to those reported in the existing literature on whole individuals and organs from different fish species in contaminated and uncontaminated freshwater and marine environments. In order to compare published REEs concentrations on a wet weight (WW) basis to that of this study on a dry weight (DW) basis, concentrations were converted to  $\mu\text{g.kg}^{-1}$  DW before comparison (Wang et al., 2022).

2.5.2. *Normalization of REEs concentrations and calculation of anomalies and ratios*  
 Data are represented as Median and Median Absolute Deviation (MAD) for  $\sum$ REEs (sum of all REEs concentrations),  $\sum$ LREEs,  $\sum$ MREEs, and  $\sum$ HREEs concentrations ( $\mu\text{g kg}^{-1}$  DW) in the target organs. MAD is a robust statistical measure of variability with little sensitivity to outliers (Leys et al., 2013). Concentrations were normalized to the Post-Archean Australian Shale (PAAS; Pourmand et al., 2012) in order to mask the Oddo-Harkins effect that reflects the strongly variable natural abundances between elements with odd and pair atomic numbers (Arienzo et al., 2022). This allows to better visualize and quantify possible anomalies in REEs patterns reflecting REEs enrichments or depletions. Anomalies were quantified for La (1;  $La_{PAAS}/La_{PAAS}^*$ ), Ce (2;  $Ce_{PAAS}/Ce_{PAAS}^*$ ) and Eu (3;  $Eu_{PAAS}/Eu_{PAAS}^*$ ) by interpolation using neighboring elements, from the following equations:

$$(1) \frac{La_{PAAS}}{La_{PAAS}^*} = \frac{La_{PAAS}}{Nd_{PAAS} + 3 \times (Pr_{PAAS} - Nd_{PAAS})} \quad (\text{Lawrence et al., 2006})$$

$$(2) \frac{Ce_{PAAS}}{Ce_{PAAS}^*} = \frac{Ce_{PAAS}}{Nd_{PAAS} + 2 \times (Pr_{PAAS} - Nd_{PAAS})} \quad (\text{Lawrence et al., 2006})$$

$$(3) \frac{Eu_{PAAS}}{Eu_{PAAS}^*} = \frac{Eu_{PAAS}}{Sm_{PAAS} \times 0.5 + Gd_{PAAS} \times 0.5} \quad (\text{Alibo and Nozaki, 1999})$$

with  $REE_{PAAS}$  the PAAS-normalized concentrations of REEs and  $REE_{PAAS}^*$  the theoretical concentration of REEs were obtained by interpolation or extrapolation.

Anomalies were considered positive for values  $> 1$  and negative for values  $< 1$  (Rétif et al., 2023). Similarly, calculating the ratios between the three groups of REEs (i.e. LREEs/MREEs, LREEs/HREEs and MREEs/HREEs) allows to study the fractionation of REEs. For this purpose, REEs without pattern anomalies have been chosen to represent each group, i.e. LREEs: Pr, MREEs: Gd and HREEs: Lu.

### 2.5.3. *Statistical processing*

Statistical treatments were applied to the data using Rstudio (RStudio Team, 2021). The non-parametric Kruskal-Wallis test was used and the results were considered significantly different when p-value <0.05. The Dunn test was used for post-hoc multiple comparison tests to allow the comparison between groups that are unbalanced in terms of number of observations (Cook and Wheater, 2005; David, 2019).

### 3. Results

#### 3.1. REEs bioaccumulation for different ages and sexes

Collected individuals were between 78.8 and 193.3 g WW and between 22 and 29 cm in total length. The otolith analysis showed that they were between 2 and 7 years old. To analyse the potential influence of age on REEs bioaccumulation, collected individuals were separated into three age groups, with Group I = 2 years old (n=4), Group II = 3 years old (n=5), and Group III > 3 years (5 to 7 years old; n= 5). There was no statistically significant difference between these age groups for  $\sum$ REEs,  $\sum$ LREEs,  $\sum$ MREEs and  $\sum$ HREEs concentrations, whatever the target organ (Kruskal-Wallis, p-value > 0.05 in all cases). Nine males and five females were collected. There was no significant difference in REEs bioaccumulation between males and females, whatever the target organ and studied element (p-value > 0.05, Table 1, S1). Accordingly, for the remaining of the manuscript, results are based on the whole data set without discriminating individuals according to their age or sex.

#### 3.2. REEs concentrations and contribution in the target organs

Median values for  $\sum$ REEs were  $213 \pm 49.8 \mu\text{g kg}^{-1}$  DW for liver,  $119 \pm 77.5 \mu\text{g kg}^{-1}$  DW for gills,  $57.7 \pm 25.5 \mu\text{g kg}^{-1}$  DW for kidneys, and  $4.38 \pm 1.20 \mu\text{g kg}^{-1}$  DW for muscles (Figure 2). Significant differences were observed in the REEs bioaccumulation between some target organs, depending on REEs groups (Kruskal-Wallis, p-value < 0.05).

Liver generally exhibited the highest values for  $\sum$ REEs,  $\sum$ LREEs, and to a lesser extent for  $\sum$ MREEs, followed by gills and kidneys. Gills displayed significantly (Kruskal-Wallis, p-value < 0.05) higher  $\sum$ HREEs concentrations than the other target organs. Muscles showed the lowest concentrations for all considered REEs groups. Overall,  $\sum$ LREEs had the highest concentrations, followed by  $\sum$ MREEs and then  $\sum$ HREEs for all organs (Figure 2).



The distribution of the different groups of REEs among the target organs, as represented by their relative contributions to  $\Sigma$ REEs, highlights that LREEs represented the major REEs fraction in all target organs (Figure 3). In all target organs, Ce (35% to 41%) was the dominant REEs, followed by La (18% to 29%), and Nd (18% to 21%). Liver displayed the lowest HREEs (1%) contribution to  $\Sigma$ REEs, compared to kidneys (5%), gills (6%), or muscles (9%). The lowest contributions to  $\Sigma$ REEs occurred for Tm (from 0.03% to 0.2%) and Lu (from 0.03% to 0.3%).

### 3.3. REEs patterns and anomalies for target organs

The method proposed by Lawrence et al. (2006) for the identification of anomalies in the REEs patterns applied to the present dataset on PAAS-normalised REEs in fish tissues (Figure 4) suggests that there are clear positive La anomalies in liver, kidneys and gills of most individuals studied. The median values of calculated La anomalies ranged from 1.03 (muscles) to 1.68 (liver), and were greater than 1.20 in all liver samples, in 54% of gills, in 79% of kidneys and only in 21% of muscles samples (Figure 5). Regarding Ce anomalies, a significant difference was observed between the liver ( $0.937 \pm 0.034$ ), which showed a negative anomaly, and the gills ( $1.07 \pm 0.119$ ), which showed a positive anomaly in *Solea solea*. Of all the individuals considered, 93% of livers showed negative anomalies and 77% of gills showed positive anomalies. The data tend to suggest positive Eu anomalies in liver ( $1.14 \pm 0.064$ ), gills ( $1.14 \pm 0.069$ ) and kidneys ( $1.18 \pm 0.051$ ), and a negative Eu anomaly in muscles ( $0.949 \pm 0.020$ ), yet the differences in Eu anomalies in the different organs were not statistically different ( $p$ -value $<0.05$ ).

There was a strong partitioning between LREEs and HREEs with a median Pr/Lu ratio of  $8.42 \pm 3.66$  for the liver, compared to ratios for the gills ( $1.92 \pm 0.55$ ), kidneys ( $2.15 \pm 0.29$ ), and muscles ( $0.744 \pm 0.15$ ). A significant difference was observed for the partitioning of LREEs and MREEs, reflected by the positive Pr/Gd ratio of the liver ( $1.39 \pm 0.10$ ) and the negative

Pr/Gd ratios of the other organs (Figure 5; Supplementary Table 1). The MREEs and HREEs partitioning as expressed by Gd/Lu ratios showed significant differences between liver ( $6.26 \pm 2.50$ ), gills ( $2.31 \pm 0.20$ ) and muscles ( $0.990 \pm 0.13$ ). Kidneys ( $3.44 \pm 0.70$ ) showed intermediate Gd/Lu ratios between liver and gills.

## 4. Discussion

### 4.1. Age and sexual influences on REEs bioaccumulation

The current dataset does not show age- or sex-related differences in REEs concentrations in *Solea solea* which is consistent with results reported for black triggerfish (*Melichthys niger*; Hauser-Davis et al., 2023). However, these results contrast with previous work, on other marine or freshwater fish species, that suggested clear differences between life stages (Mayfield and Fairbrother, 2015; MacMillan et al., 2017; Lortholarie et al., 2020) as well as sex (MacMillan et al., 2017; Lortholarie et al., 2020). These contrasting findings may be attributed to the fact that these authors have studied different life stages or a wider range of ages (MacMillan et al., 2017; Lortholarie et al., 2020), whereas the present work addresses only adult individuals of *Solea solea*. Furthermore, there is a stronger sexual dimorphism in other fish species implying differences in growth, feeding behavior, lipid content, etc. (e.g. eel; Lortholarie et al., 2020), whereas sexual dimorphism is weak in *Solea solea* (James et al., 1986). The possibility that other external environmental factors may influence the presence/absence of sex- or age-related REEs accumulation cannot be ruled out.

### 4.2. Organ-specific concentrations and patterns

The bioaccumulation of REEs displays significant differences within distinct organs (organotropism), showing the highest concentrations in liver (e.g. REEs, LREEs and MREEs; Figure 2), except for HREEs in gills.

This present study shows that *Solea solea* individuals accumulated more LREEs (Ce > La > Nd) than MREEs or HREEs (Figure 3). These results reflect to some extent the natural abundance of REEs in the Earth's crust, where LREEs are naturally more abundant than HREEs (Bru et al., 2015).

REEs concentrations in organs of *Solea solea* from the WGMP have been compared to those reported in the existing literature (Figure 6). The observed REEs concentrations in organs of *Solea solea* were overall similar in magnitude to those reported in the existing literature (MacMillan et al., 2017; Lortholarie et al., 2021; Squadrone et al., 2022).

#### 4.2.1. Organotropism

The  $\Sigma$ REEs organotropism of *Solea solea* followed a general pattern: liver > gills > kidneys > muscles (Figure 2). In the Bay of Biscay, concentrations in liver, gills, kidneys, and muscles in male silver European eel from the Loire Estuary (Lortholarie et al., 2021), ~200 km to the north of our study site, and the resulting organotropism were rather similar to those measured in *Solea solea* collected in the WGMP. The REEs concentrations in liver of Brook Trout (*Salvelinus fontinalis*), a freshwater fish from the eastern Canadian Arctic (MacMillan et al., 2017), are similar in magnitude as those in liver of *Solea solea* from the WGMP. Hauser-Davis et al. (2023) reported  $\Sigma$ REEs concentrations in black triggerfish (*Melichthys niger*), a demersal species from a pristine area in the South Atlantic Ocean, which were roughly three to five-times lower than ours results. Despite large differences in concentration, the organotropism found by these authors is close to that we observed in *Solea solea*. Similarly, Squadrone et al. (2022) reported  $\Sigma$ REEs concentrations in spotted dogfish (*Scyliorhinus stellaris*), a chondrichthyes benthic species, obtained from an aquatic park, as follows: liver ( $30 \mu\text{g kg}^{-1}$  WW) > muscle ( $15 \mu\text{g kg}^{-1}$  WW) > kidney ( $13 \mu\text{g kg}^{-1}$  WW). If the concentrations in liver and kidneys are in good agreement with our data,  $\Sigma$ REEs in muscle of spotted dogfish were clearly higher. The latter observation strongly supports the need for more systematic research on REEs in natural environments to allow for advanced comparison.

Hauser-Davis et al. (2023) suggested that the observed REEs concentrations and the resulting organotropism may result from dietary exposure rather than contact with the sediment. In fact, previous works have reported that REEs in pelagic fish species tend to be lower than those in

benthic species, attributed to the greater proximity of the latter to sediments containing relatively high REEs due to their mineral content (Mayfield and Fairbrother, 2015; Wang et al., 2019). Given that *Solea solea* is a benthic fish mainly feeding on benthic macrofauna (Lagardère, 1987), this may explain higher concentrations in organs of *Solea solea* from the WGMP than in those of black triggerfish (*Melichthys niger*; Hauser-Davis et al., 2023).

The general scarcity of published data on REEs in fish organs (e.g. liver, kidneys, gills) and the disparity of organs and elements targeted in these pioneer studies (MacMillan et al., 2017; Squadrone et al., 2020; Souza et al., 2021; Lortholarie et al., 2021; Squadrone et al., 2022; Hauser-Davis et al., 2023), limit the comparison of REEs in these organs between different species and geographical regions, clearly suggesting the need for more systematic monitoring of REEs in different fish species worldwide (Figure 6).

#### 4.2.2. Organ-specific patterns

The distribution of the different REEs groups, as well as that of the elements within their group, differ between the target organs (Figure 3), which implies differences in PAAS-normalized patterns of the various organs.

The organs exhibited different patterns of REEs (Figure 4), potentially reflecting different absorptive pathways through the organism, or differences in regulatory and excretory systems.. Liver exhibits LREEs ~1000 times lower than the respective elements in the normalizing reference material PAAS, whereas PAAS-normalised HREEs in liver are up to one order of magnitude lower than PAAS-normalised LREEs, as previously observed in Brook Trout liver by MacMillan et al. (2017).

The strong partitioning between LREEs and HREEs for the liver, compared to ratios for the other target organs, may suggest a different metabolism in terms of managing the detoxification and/or elimination capacities of HREEs in the liver compared to other organs (Figure 5). This

pattern clearly contrasts with that of kidneys and gills, where PAAS-normalised MREEs tend to be higher than those of LREEs and HREEs, the latter appearing as even more depleted than the LREEs (Figure 4).

The relative depletion of HREEs in all organs studied, except for muscle (Figure 4) may be attributed to higher availability of LREEs reflecting greater solubility (Pratas et al., 2017). However, unlike the liver, muscle is not an organ of metabolism/excretion, implying differences in metabolic levels between the target organs due to their also very different physiological roles. Moreover, this may have repercussions on the management of metallic elements, which is different for each organ. These differences could explain the contrast with the more homogeneous REEs pattern in muscle, which is relatively similar to a classic seawater pattern (Elderfield et al., 1990). Accordingly, the bioaccumulation and the partitioning between REEs groups can be attributed to uptake pathways and function of the different organs, such as transfer, detoxification or storage organs.

Positive La anomalies in seawater have been attributed to processes in brackish waters along estuarine salinity gradients (Lawrence and Kamber, 2006). The observed differences in La anomalies in the liver, gills and kidneys of *Solea solea* may therefore reflect REEs partitioning by the different organs implied in the transfer (i.e. gills) and detoxification (i.e. liver and kidneys). In addition, the kidneys allow osmotic regulation of the organism (Takvam et al., 2021), which could also contribute to the presence of these positive anomalies in La. Interestingly, similar or even slightly greater La anomalies also occurred in liver of European eel (calculated from supplementary data in Lortholarie et al., 2021).

MacMillan et al. (2017) have reported negative Ce anomalies in various aquatic organisms, including brook trout liver, similar to that observed in *Solea solea* liver. These authors potentially attribute these anomalies to a change in the degree of oxidation of Ce from Ce<sup>3+</sup> to Ce<sup>4+</sup>, implying a lower solubility of the latter. However, the present dataset shows a positive

Ce anomaly in the gills of *Solea solea*, an organ that has been seldomly studied in fish (Hauser-Davis et al., 2023; Lortholarie et al., 2021; Souza et al., 2021). Hence, future work should aim to understand the mechanisms of REEs transfer and organ-specific accumulation through both, the transfer (i.e. gills) and detoxification (i.e. liver and kidneys) organs.

The PAAS-normalized patterns provided slight positive Eu anomalies in the studied fish, which is in good agreement with previous studies on a large variety of biological samples, including terrestrial plants, freshwater fish, birds, marine invertebrates, and mammals (MacMillan et al., 2017). The absence of significant differences between target organs in our dataset might suggest that Eu anomalies cannot result from specific functions of each organ.

Further work on the respective roles of (i) the regional geological background, and (ii) physiologically induced changes of REEs patterns in different biological species is necessary to reliably define reference patterns for biological samples and then identify anomalies that reflect natural environmental processes and/or emerging anthropogenic REEs sources.

#### *4.2.3. Potential controls on organ-specific distribution*

For HREEs, the highest concentrations in the gills are consistent with observations in European eel at the yellow eel stage (Lortholarie et al., 2021). The fact that the highest concentrations of HREEs occurred in gills, rather than in liver, probably reflects the relatively low proportion of HREEs in liver, compared to the other organs (i.e. gills, kidneys, and muscles). This may potentially be explained by the direct REEs uptake pathway (i.e. respiration via the gill epithelium) together with relatively stronger fixation of the HREEs by the replacement of calcium in the epithelium (Martin and Richardson, 1979; Figueiredo et al., 2018). In fact, HREEs are efficient antagonists of calcium absorption (Evans, 1983) and marine fish mainly absorb calcium through the gills (Flik et al., 1995; Baldisserotto, 2019).

Numerous studies have shown that metals such as Cu, Zn, and Pb concentrate mostly in the liver and kidneys (Fernandes et al., 2008; Shahjahan et al., 2022) because these organs are the primary sites of metal sequestration or detoxification (e.g. Pannetier et al., 2016). Given that liver is an organ responsible for the detoxification of metals (Hauser-Davis et al., 2023), this might explain higher concentrations of REEs in liver of *Solea solea* compared to the other target organs.

The relatively higher accumulation of REEs in different detoxification organs (i.e. liver and kidneys) compared to muscle could be attributed to the higher metabolic activity of those organs compared to muscle (Serra et al., 1993; Canli et al., 1998), as well as lower absorption and/or binding power of muscle (Uluturhan and Kucuksezgin, 2007; Pan et al., 2022). This explains that the REEs concentrations in muscle were close to or even below detection limits, in agreement with previous works, reporting that REEs occurred at ultra-trace levels or were not detected in fish muscles (Mayfield and Fairbrother, 2015; Yang et al., 2016; MacMillan et al., 2017, Lortholarie et al., 2020). Considering that (i) human consumption of *Solea solea* usually is restricted to muscle and (ii) the general trophic dilution along the aquatic food chain (Santos et al., 2023), one would expect potential health risks due to consumption of *Solea solea* from the WGMP to be very low.

## 5. Conclusion

This study presents the first data on REEs contents in different organs of the common sole (*Solea solea*) from the West Gironde Mud Patch, allowing the establishment of a baseline for future investigations in this area and at a greater scale, in the Bay of Biscay (NE Atlantic Ocean). As such, this work contributes to fill the general knowledge gaps in the assessment of the accumulation of REEs in marine fish, which is still largely under-documented. The REEs concentrations in adult soles were within the range of values previously reported for various fish freshwater and marine species worldwide, and did not show any significant age or sex-



related difference. Comparison with the few existing publications on REEs organotropism confirmed relatively greater REEs accumulation in liver and lowest in muscles, consistent with literature on REEs. There are clear differences in REEs distribution patterns between target organs, suggesting internal distribution processes favoring HREEs accumulation in gills, compared to LREEs. The global organotropism of REEs in *Solea solea* exhibited a consistent trend, with the highest accumulation found in the liver, followed by the gills, kidneys, and muscles. The role of exposure, uptake pathways and physiological processes in the redistribution of REEs in fish needs to be further explored. The approach of identifying anomalies in the different organs using PAAS-normalised REEs patterns seems to reflect different organ functions. There is a general need for more systematic monitoring of REEs in different species of fish (and other marine organisms) worldwide to establish reliable (i) REEs baselines (ii) factors (e.g. exposure pathway, physiological processes, species-specific responses) controlling uptake and bioaccumulation of REEs and (iii) environmental quality standards for wildlife and especially coastal aquatic organisms, which may be needed as biomonitors for the emerging REEs contamination of coastal systems expected in the near future.

## **CRedit authorship contribution statement**

Maëva LABASSA : Investigation, Formal analysis, Data curation, Visualization, Writing - Original Draft, Writing - Review & Editing

Clément PERETO : Formal analysis, Data curation, Visualization, Writing - Original Draft, Writing - Review & Editing

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## **Declaration of competing interest**

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

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