



Review

Global natural concentrations of Rare Earth Elements in aquatic organisms: Progress and lessons from fifty years of studies

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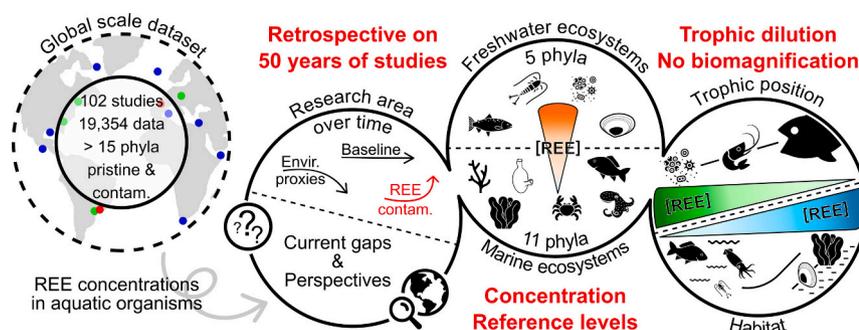
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HIGHLIGHTS

- Need for an overview on the natural REE concentrations in aquatic organisms
- Knowledge on control factors and reference levels for REE accumulation in organisms
- Retrospective on 50 years of REE concentration studies in aquatic organisms
- 16 reference levels for REE concentrations in freshwater and marine phyla
- Influence of habitat and trophic dilution of natural REEs on a global scale

GRAPHICAL ABSTRACT



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ABSTRACT

Rare Earth Elements (REEs) consist of a coherent group of elements with similar physicochemical properties and exhibit comparable geochemical behaviors in the environment, making them excellent tracers of environmental processes. For the past 50 years, scientific communities investigated the REE concentrations in biota through various types of research (e.g. exploratory studies, environmental proxies). The extensive development of new technologies over the past two decades has led to the increased exploitation and use of REEs, resulting in their release into aquatic ecosystems. The bioaccumulation of these emerging contaminants has prompted scientific communities to explore the fate of anthropogenic REEs within aquatic ecosystems. To achieve this, it is necessary to determine the natural concentration levels of REEs in aquatic organisms and the factors controlling REE dynamics. However, knowledge gaps still exist, and no comprehensive approach currently exists to assess the REE concentrations at the ecosystem scale or the factors controlling these concentrations in aquatic organisms.

Based on a database comprising 102 articles, this study aimed to: i) provide a retrospective analysis of research topics over a 50-year period; ii) establish reference REE concentrations in several representative phyla of aquatic ecosystems; and iii) examine the global-scale influences of habitat and trophic position as controlling factors of REE concentrations in organisms. This study provides reference concentrations for 16 phyla of freshwater or marine organisms. An influence of habitat REE concentrations on organisms has been observed on a global scale. A trophic dilution of REE concentrations was highlighted, indicating the absence of biomagnification. Lastly, the retrospective approach of this study revealed several research gaps and proposed corresponding perspectives to

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address them. Embracing these perspectives in the coming years will lead to a better understanding of the risks of anthropogenic REE exposure for aquatic organisms.

1. Introduction

Rare Earth Elements (REEs) are a group of 17 elements, consisting of scandium (Sc), yttrium (Y), and the 15 elements of the lanthanide series: lanthanum (La), cerium (Ce), praseodymium (Pr), neodymium (Nd), promethium (Pm), samarium (Sm), europium (Eu), gadolinium (Gd), terbium (Tb), dysprosium (Dy), holmium (Ho), erbium (Er), thulium (Tm), ytterbium (Yb), and lutetium (Lu). Only Pm does not have a stable isotope in the environment. In the Upper Continental Crust (UCC), REEs are relatively abundant (e.g. Taylor and McLennan, 1985; Viers et al., 2009), with a factor of 200 separating the most abundant REE (Ce; 64 mg.kg⁻¹) from the least abundant (Lu; 0.32 mg.kg⁻¹). Some REEs are naturally more abundant than commonly studied trace metal elements (TMEs) such as lead (20 mg.kg⁻¹), uranium (2.8 mg.kg⁻¹), or antimony (0.2 mg.kg⁻¹). The natural abundance of REEs is characterized by: i) a higher abundance of REEs with even atomic numbers compared to those with odd atomic numbers; and ii) a higher abundance of REEs with low atomic numbers compared to those with high atomic numbers. Thus, the natural abundance of REEs follows the Oddo-Harkins rule (e.g. Arienzo et al., 2022 and references therein), and these elements can be classified into three groups based on their atomic masses (e.g. Haque et al., 2014; Piarulli et al., 2021): the light REEs (LREEs) from La to Nd; the medium REEs (MREEs) from Sm to Gd; and iii) the heavy REEs (HREEs) from Tb to Lu.

REEs have similar physicochemical properties due to their close electronic configurations, which give them a coherent chemical behavior in the environment as a group of elements (Elderfield et al., 1990 and references therein). However, slight differences in chemical reactivity can be observed in the environment due to minor differences in electronic configurations. The 4f valence electron shell is gradually filled with increasing atomic number (Sonke and Salters, 2006), leading to a screening effect of atomic charge and a progressive decrease in ionic radius (i.e. lanthanide contraction). Thus, REEs exhibit slightly different behaviors from one element to another in the environment. REEs are trivalent, characterized by a highly stable +3 oxidation state, with two exceptions: Ce and Eu. Europium (Eu³⁺) tends to be divalent in reducing environments (Eu²⁺). Conversely, Ce³⁺ tends to lose its valence electron to become Ce⁴⁺ in oxidizing environments. In oxidizing oceanic environments, Ce⁴⁺ precipitates as CeO₂, creating fractionation of REEs in the water column. These fractionations have been used as proxies for ocean oxygenation conditions in paleoceanology (German and Elderfield, 1990). More generally, the lanthanide contraction and the unique behavior of Ce and Eu under redox conditions have been extensively explored in the literature to understand environmental processes (e.g. mechanical erosion, chemical weathering of rocks, diagenesis, reactivity in the water column; Aubert et al., 2001; Elderfield et al., 1990; Fedele et al., 2008; Laveuf and Cornu, 2009; Smrzka et al., 2019). Therefore, REEs serve as excellent geochemical tracers of these processes, explaining the numerous geochemical studies focused on these elements since the 1960s (e.g. Arienzo et al., 2022; Migaszewski and Gałuszka, 2015; Piper, 1974 and references therein).

The last twenty years, the exploitation of REE resources has intensified due to their use in many technologies (e.g. renewable energy, multimedia, petroleum industry, medicine; Balaram, 2019). This increased usage has led many authors to investigate human exposure levels to REEs and the associated toxicity (e.g. Benedetto et al., 2018; Pagano et al., 2015, 2019). Furthermore, this increased usage has led to more pronounced releases of REEs into the aquatic environment (e.g. Brito et al., 2018; Hatje et al., 2016; Kulaksız and Bau, 2013). These anthropogenic REEs are now considered as emerging contaminants and are observed in both freshwater and marine ecosystems. These emerging

contaminants have multiple anthropogenic sources (e.g. mining operations, urban discharges, petroleum industry, traffic, agriculture; Delgado et al., 2012; Kulaksız and Bau, 2011; Lerat-Hardy et al., 2019, 2021; Olmez et al., 1991). Several authors have investigated fluxes and geochemical behaviors of anthropogenic REEs (e.g. Kulaksız and Bau, 2007, 2013; Lerat-Hardy et al., 2021; Pereto et al., 2023; Tranchida et al., 2011). In recent years, an increasing number of studies have highlighted the presence of these contaminants in aquatic organisms (e.g. Castro et al., 2023; Gaudry et al., 2007; Ma et al., 2019; Merschel and Bau, 2015) and have investigated the toxicity of these elements (e.g. Hanana et al., 2017; Herrmann et al., 2016; Pagano et al., 2016; Parant et al., 2019). Understanding the fate of these contaminants in the total environment represents a scientific challenge and raises major concerns about the associated health and environmental risks.

A thorough understanding of the natural biogeochemical cycles of REEs is required to solve this challenge. Understanding these cycles involves characterizing the REE concentrations in the environment, both in abiotic and biological compartments. On a large scale, some authors have proposed reference REE concentrations for abiotic compartments such as river suspended particulate matter (SPM; Viers et al., 2009), river waters and sediments (Bayon et al., 2015; Gaillardet et al., 2003), or even ocean seawaters and SPM (Turekian, 2010 and references therein). Regarding aquatic organisms, numerous studies have investigated the natural REE concentrations (e.g. Kano et al., 2002; Lobus et al., 2019; Sena et al., 2022; Sholkovitz and Shen, 1995). However, no study proposes REE reference concentrations for aquatic organisms. These reference concentrations in uncontaminated aquatic organisms are one of the keys to better understanding the global fate of REEs. Furthermore, knowing these uncontaminated reference concentrations would facilitate the observation of cases of REE contamination.

Several studies have investigated the fate of REEs along food webs and have notably demonstrated trophic dilution of REE concentrations and an absence of biomagnification (e.g. MacMillan et al., 2017; Rétif et al., 2024; Santos et al., 2023). Other studies have observed a strong influence of REE concentrations in the habitat (i.e. water, sediments) on species, depending on their position in the habitat (i.e. pelagic, benthic; Amyot et al., 2017; Yang et al., 2016). However, these studies have focused on local ecosystems, covering few different phyla. A global-scale perspective on REE concentrations in organisms, considering their general trophic positions and habitats, would be a relevant approach to better understand the dynamics of REEs in aquatic ecosystems.

Based on literature data, the main objective of this study is to propose reference concentrations for REEs in uncontaminated aquatic organisms, serving as tools for the scientific community to contextualize their work on a global scale. To achieve this, the study relies on a compilation of 102 articles covering 50 years of research on REE concentrations in aquatic organisms (Pereto et al., 2024), and is structured around three questions: i) What is the current spatial and temporal distribution of available data? ii) What are the reference concentrations of REEs in several phyla of freshwater and marine organisms? and iii) What relationships exist between these organisms, their trophic position, and their habitat on a global scale? Finally, special emphasis is placed on current gaps in knowledge and the associated research perspectives regarding the study of REE concentrations in aquatic organisms.

2. Materials and methods

2.1. Details of literature search strategy

The literature search was conducted multiple times on Google Scholar up until February 2023, using various combinations of the

following keywords: “REEs,” “Rare Earth,” “Rare Earth Elements,” “Anthropogenic,” “Trace Metals,” “Accumulation,” “Bioaccumulation,” “Concentration,” “Organisms,” “Freshwater organisms,” “Marine organisms,” “Species,” “Freshwater species,” and “Marine species.” A first screening was performed to exclude articles that were not relevant (e.g. laboratory experiments, materials chemistry) or did not focus on in situ sampling of aquatic organisms. A second screening was conducted to remove duplicate articles. However, four laboratory experimentation articles were included for the data of uncontaminated controls when these organisms were originally sampled from the natural environment (Andrade et al., 2020; Figueiredo et al., 2018, 2022a; Qiang et al., 1994). Ultimately, 102 articles were included in this study, and the compiled data is available in the form of a database (Pereto et al., 2024), for ensuring access to research data to the scientific community. All references to these articles are available in the database and in Supplementary Table A.1.

Compared to the bibliometric analysis by Blinova et al. (2020), which reported only 18 articles (out of 241) derived from aquatic organisms in the natural environment, we present a compilation of 102 articles, with a total of 19,354 data points. The database compiles REE concentration values for 16 phyla, represented by 36 phylogenetic classes and distributed among freshwater and marine ecosystems. 19 articles provide data for both marine and freshwater organisms, 72 for marine organisms, and 11 for freshwater organisms. These data, spanning 37 countries, cover a significant part of the globe (number of data points): Polar Oceans (n = 341), Atlantic Ocean (n = 7205), Pacific Ocean (n = 5530), Europe with Mediterranean and Baltic Seas (n = 3085), Asia (n = 1020), America (n = 1952). It is important to note that this database is not intended to be exhaustive, but we have estimated that the database is representative of studies conducted on REE concentrations in aquatic organisms.

2.2. Database structure

The database (Pereto et al., 2024) includes articles where at least one REE has been analyzed in at least one aquatic organism. The REE concentrations in abiotic compartments (e.g. water, sediments) from these studies have also been included in the database. The REE concentrations, or the sum of REE concentrations (\sum REEs), have been categorized based on several criteria: i) the bibliographic reference of the article and its publication year; ii) the name of the studied species; iii) the phylum of the studied species (e.g. mollusks, arthropods, chlorophytes); iv) the general trophic position (e.g. primary producer, primary consumer); v) the general habitat (i.e. benthic, pelagic); vi) the ecosystem (i.e. marine, freshwater); vii) the analyzed tissue (e.g. whole organism, muscle, leaf); viii) the presence or absence of REE contamination due to anthropogenic source(s); ix) the country and geographic area of sampling (e.g. France, Brazil, North Atlantic Ocean, China Sea); x) the GPS coordinates associated with the sampling sites. When the species name is not specified by the authors, the phylum or the phylogenetic classes has been used. Concerning phyla, two exceptions may be given: phytoplankton and zooplankton. However, for the sake of simplicity, we will also refer to them as phyla. The phyla of the species were obtained from the World Register of Marine Species (WoRMS Editorial Board, 2023) when this information was not provided by the authors. The trophic position and habitat of the species were obtained from FishBase (Froese and Pauly, 2022), SeaLifeBase (Palomares and Pauly, 2022) or from specific literature (for some freshwater species) when this information was not available. The GPS coordinates of the sampling sites were determined as accurately as possible using Google Maps when they were not provided.

2.3. Data processing

2.3.1. Data distribution analysis

The entire database has been standardized to ng.g^{-1} for biological and sedimentary matrices, and $\mu\text{g.L}^{-1}$ for freshwater and seawater

concentrations. For biological matrices, the database provides REE concentrations based on dry weights (DW) and wet weights (WW) for, respectively, 99 % and 1 % of the used database. Due to the wide ranges of concentrations observed in aquatic organisms, no significant difference was observed between the concentrations expressed in DW and WW (Kruskal test; p-value > 0.05). Hence, both types of concentrations were considered without distinction in the rest of the study (the distinction between DW and WW is available in the database; Pereto et al., 2024). Concentrations expressed in moles have also been converted, except for the molar concentrations of \sum REEs, which were not considered in this study. Furthermore, no \sum REEs calculation was performed if the authors did not provide these data in their work.

The distribution of available data was assessed according to: i) the analyzed REEs; ii) the publication years of the articles; iii) the geographical origin of the data (cartography); and iv) the phylum of the studied species. The temporal evolution of the number of available data was conducted across four time periods: before 2000; 2000–2009; 2010–2019; and since 2020. The cartography of sampling sites was conducted using a GIS software (Geographic Information System; ArcGIS). The data from these studies were represented based on: the major geographical zones; the sampling ecosystem; the presence or absence of REE contamination; and the phylum of the studied species (only for REE uncontaminated sites). Regarding the distinction between the presence and absence of REE contaminations, any sample for which the authors considered the presence of contamination in one or more REEs was classified as “contaminated”.

2.3.2. Analysis of REE concentration levels in organisms

A correlation matrix was generated using the Spearman method (Hauke and Kossowski, 2011 and references therein) for the concentration data of the 14 REEs. The data used for this matrix corresponded to all uncontaminated concentration data of REEs (all phyla, all tissues, all environments combined). The correlation coefficients (r) were considered significant for p-values < 0.001 ($\alpha = 0.1\%$).

At a global scale, reference levels of natural REE concentrations were determined for uncontaminated environments as following: i) whole organisms were used to avoid potential heterogeneity introduced by large differences between analyzed tissues; ii) representative phyla from freshwater and marine ecosystems were considered; and iii) a minimum of 5 observations per phylum were considered. Two exceptions were made regarding whole organisms: i) the phylum of cnidarians (i.e. exclusively hard corals in the database) was considered for their calcareous skeletons; ii) mollusks with shells (e.g. bivalves, snails), included in the phylum Mollusca, were considered for their entire soft tissues (i.e. without the shells).

All the data used in this study come from published articles with possible data subject to caution due to the evolution of analytical techniques, the consideration or not of interferences during REE analyses, or too significant detection limits. Consequently, a robust statistical analysis method has been adopted to minimize these potential biases as much as possible, without having to make a subjective judgment on published data. Estimates of central tendency for REE concentrations were calculated using medians to minimize the influence of extreme values (i.e. outliers). Median Absolute Deviation (MAD) was chosen as a measure of dispersion because it is a robust statistical measure of variability, less sensitive to outliers (Leys et al., 2013). The sums of median concentrations and MAD (\sum REEs_{med.} \pm \sum MAD) were also calculated to provide a simple account of the concentration range of the different phyla studied. Statistical tests for median comparisons were performed using the non-parametric Kruskal-Wallis test (McKight and Najab, 2010) based on ranks, with a significance threshold (α) of 5 %. Post-hoc multiple comparisons were conducted using the Dunn test (Cook and Wheeler, 2005; David, 2019), which is suitable for unbalanced groups with different sample sizes. Due to a large number of comparisons, a Bonferroni correction (Armstrong, 2014) was applied to adjust the p-values and minimize potential Type I errors (false positives).

All statistical and data processing procedures were conducted using R (RStudio; RStudio Team, 2021).

3. Results and discussion

3.1. Retrospective on the available data of REE concentrations in organisms

3.1.1. Data distribution by REE

The entire database compiled 19,354 data of REEs, with 19,013 concentrations of individual REE and 341 data of \sum REEs. The number of available data per REE (Fig. 1A) shows a factor of 2 between the most frequently detected REE (La) and the least detected REE (Tm). Except for La, even-numbered REEs (i.e. Ce, Nd, Sm, Gd, Dy, Er, Yb) are more frequently detected than their odd-numbered neighbors (i.e. Pr, Eu, Tb, Ho, Tm, Lu), and LREEs are more frequently detected than HREEs. This pattern of analysis frequency by element corresponds to the natural abundance of REEs in the environment, as demonstrated with the comparison with the Upper Continental Crust (UCC; Fig. 1B; Taylor and McLennan, 1985) and strong positive correlations ($r = 0.86$; p -value < 0.001), except for only Tm that appears to be under-represented relative to its natural abundance (see reasons below).

Since their introduction in the 1980s, Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) has become the preferred technique for analyzing trace elements, and many authors have focused on its development (Thomas, 2013 and references therein), especially for the quantification of REEs (e.g. Wysocka, 2021; Zawisza et al., 2011; Zhu, 2020). In the database, 91 % of the REE analyses were performed using ICP-MS. Over the past 40 years, the development of ICP-MS has addressed various analytical challenges such as spectral and non-spectral interferences (e.g. Lum and Leung, 2016; Wysocka, 2021). Furthermore, the progressive lowering of detection limits has facilitated the quantification of the least abundant REEs (i.e. Tb, Ho, Tm, Lu). These technological advancements have enabled the quantification of increasingly lower concentrations, partially explaining the observed correlation between the number of analyses and the natural abundance of REEs (Fig. 1B). In addition to low environmental concentrations, the frequent use of Tm as an internal standard during REE analysis by ICP-MS (e.g. Barrat et al., 2022; Le Goff et al., 2019; Merschel and Bau, 2015; Ponnurangam et al., 2016) may explain the lower number of data points for this element and the deviation observed in the correlation. These initial observations highlight that there are still gaps in the amount of information available for each REE.

3.1.2. Temporal distribution

Of the 102 articles published over the past 50 years (1973–2023), it

is possible to highlight different research topics that have required the analysis of REEs in aquatic organisms. Only four of these studies were published before the year 2000, 15 articles between 2000 and 2009, 44 articles between 2010 and 2019, and 39 articles have been published since 2020 (i.e. 2020–Feb. 2023). This temporal distribution of the number of articles is consistent with the bibliometric analysis by Blinova et al. (2020) for the period 1991–2019. To better understand the temporal evolution of the different research topics, the available data in the database has been represented by phylum for the four time periods (Fig. 2).

A quarter of the studies provide concentration levels for one or more REEs, along with other TMEs. These studies have a multi-element approach (e.g. baseline, exploratory, natural sources, non-REE anthropogenic sources), without specifically targeting REEs. These articles are evenly distributed over the past 50 years and provide REE concentrations in some phyla of aquatic organisms for the first time (e.g. Bustamante and Miramand, 2005; Escobedo Mondragón et al., 2021; Hou and Yan, 1998; Squadrone et al., 2016). Before 2000, the study of Hou and Yan (1998) provided the first information on REE concentrations in chlorophytes ($n = 34$), ochrophytes ($n = 72$), and rhodophytes ($n = 62$). In the 2000–2009 period, data from Ichihashi et al. (2001) and Pernice et al. (2009) informed us about the REE concentrations in cephalopod mollusks ($n = 310$ out of 400 mollusk data in the period). However, until 2010, only eight phyla had been studied (Fig. 2). Since 2010, studies have provided the first information on REE concentrations in sponges ($n = 187$; Figueiredo et al., 2021; Orani et al., 2022), annelids ($n = 238$; Parisi et al., 2017), and phytoplankton ($n = 287$; Dang et al., 2023; Lobus et al., 2021; Strady et al., 2015). These studies partly explain the diversification of studied phyla in the last 20 years. However, data regarding, for example, echinoderms ($n = 1$; MacMillan et al., 2017), tunicates ($n = 70$; Parisi et al., 2017), or bryophytes ($n = 56$; Pratas et al., 2017) are still under-represented, with only one study per phylum. However, this knowledge is necessary to better understand the dynamics of REEs within ecosystems, particularly between trophic levels. These studies which aims to define REE concentration levels in new species and phyla, are therefore essential to achieve this goal.

Another quarter of the studies in the database focused on REEs for their unique electronic, physical, and chemical properties. These studies relied on the available knowledge regarding the geochemical behavior of REEs and their reactivity in the environment to apply it to aquatic organisms. Due to slight differences in electronic configurations, REEs exhibit slight variations in geochemical behavior and undergo fractionation processes that reflect environmental processes (e.g. Elderfield et al., 1990; Louis et al., 2020; Sholkovitz, 1995). Therefore, these studies have investigated REE concentrations in aquatic organisms as a proxy of environmental processes in aquatic environments (e.g.

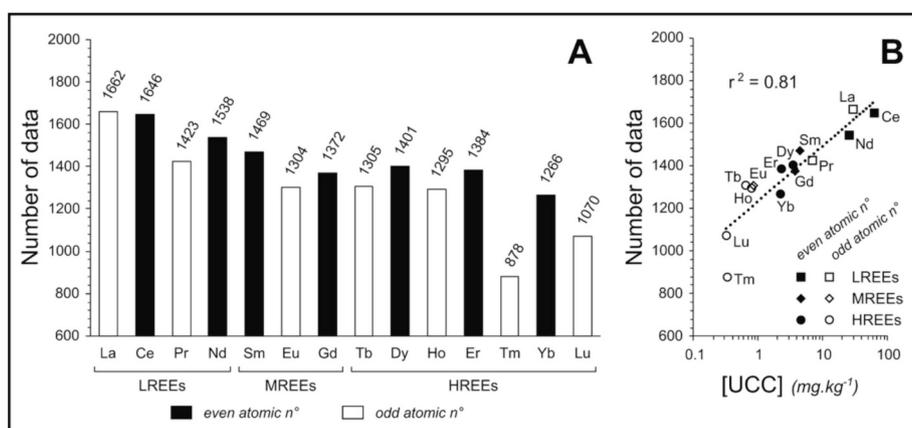


Fig. 1. A) Number of available concentration data for each REE. B) Number of available concentration data compared to the concentration of each REE in the upper continental crust (UCC; mg.kg^{-1} ; data from Taylor and McLennan, 1985). Logarithmic correlation ($r^2 = 0.81$). REEs are classified according to their group (i.e. LREEs, MREEs, HREEs) and their atomic number (i.e. even or odd).

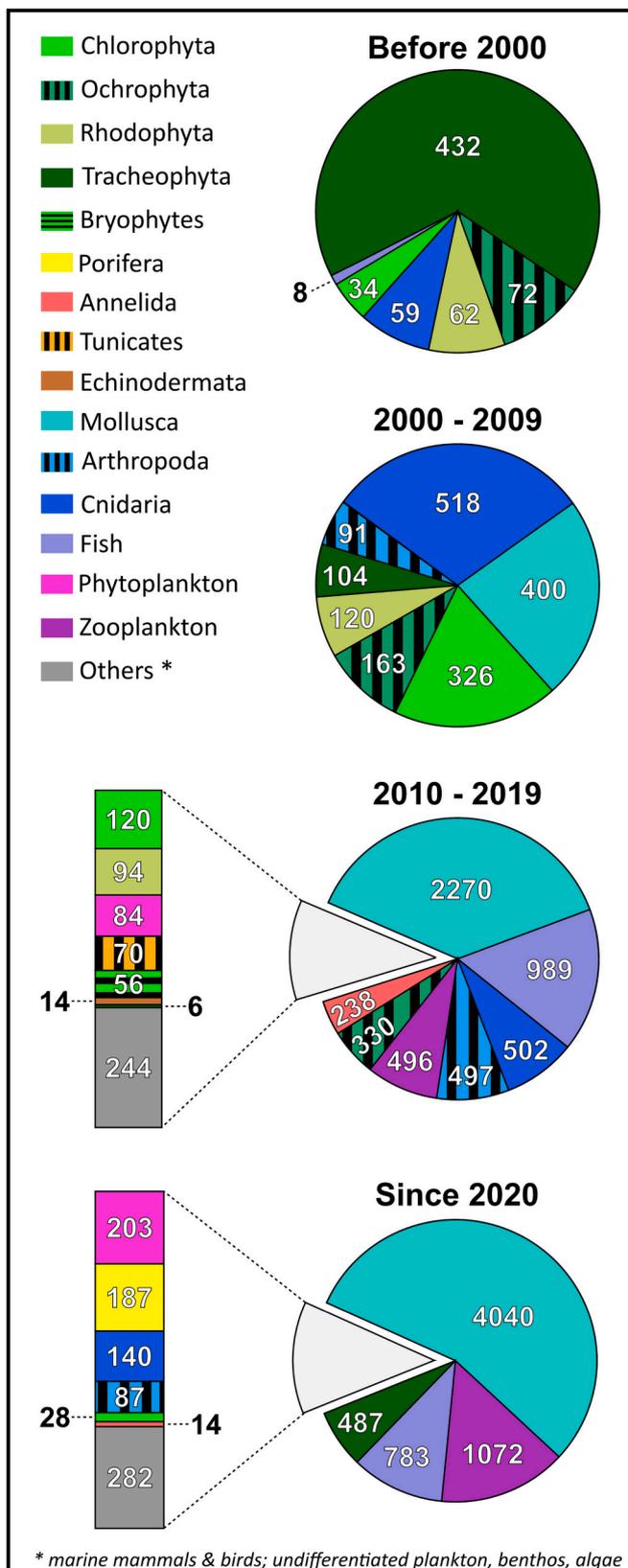


Fig. 2. Number of available concentration data in aquatic organisms according to the time period (i.e. before 2000; 2000–2009; 2010–2019; since 2020) and according to the studied organism’s phylum. The “Others” category includes data on marine mammals, seabirds, marine reptiles, and organisms for which the phylum or species group is not identifiable (e.g. algae, plankton, benthos).

physicochemical variations in water bodies, transport and dispersion of material in the critical zone; Liu et al., 2017; Sakamoto et al., 2008; Valdés-Vilchis et al., 2021). These research objectives were prominent in the periods before 2010 and appear to represent an application of knowledge from REE geochemistry to REE concentrations in organisms. Before 2000, the studies of Cowgill (1973) on tracheophytes (vascular plants; n = 432) and Sholkovitz and Shen (1995) on cnidarians (n = 59) investigated this link between biology and geochemistry. Between 2000 and 2009, these objectives accounted for 47 % of the studies (7 articles), including all the data concerning cnidarians (n = 518; e.g. Akagi et al., 2004; Fallon et al., 2002; Jupiter, 2008; Wyndham et al., 2004). In reality, all studies focusing on cnidarians in the last 50 years have examined scleractinian cnidarians (i.e. hard corals) as environmental bioarchives, for short-term (e.g. seasonality) or long-term (e.g. paleoclimate) purposes. These research objectives explain the presence of data for this phylum across the four temporal periods (Fig. 2). Bivalve mollusk shells are also used as bioarchives for studying REE dynamics in the environment (e.g. Bau et al., 2010; Mouchi et al., 2020). Since 2010, the use of REE concentrations in organisms as a proxy for environmental processes appears to be less prevalent, accounting for 25 % (11 articles) of the studies in the 2010–2019 period and only 15 % since 2020 (6 articles). In reality, this apparent decrease is linked to the appearance and increase of a new research topic.

Half of the studies in the database are related to this new research topic and are characterized by a significant increase in the available data for mollusks, zooplankton, and fishes since 2010. These topic account for 43 % (19 articles) and 67 % (26 articles) of the studies in the periods 2010–2019 and post-2020, respectively. Until recently, the study of REEs as contaminants in ecosystems was given little consideration. These elements were, for a long time, considered as non-essential, associated with low (eco)toxicological risk, and absent from anthropogenic releases (Arciszewska et al., 2022; Cotruvo, 2019). However, since the 1990s, REEs have been used as nutrients in agricultural fertilizers (Pang et al., 2002) and in the feed of farmed species (Abdelnour et al., 2019). The scientific community has thus begun to question the potential transfers and impacts of these elements on humans and the environment. The first article in the database to address this issue was conducted in China on the fish *Cyprinus carpio* (Qiang et al., 1994). The last two decades have been marked by the intensive expansion of new technologies that require the industrial-scale exploitation of REEs (Balaram, 2019). The increasing global use of REEs has resulted in the release of these emerging contaminants into the environment and the proliferation of studies over the past 20 years.

One of the questions in these recent studies is the potential transfer of anthropogenic REEs into the human food chain. These studies, particularly focused on fish muscle tissues as a potential source, have generated a large amount of data (e.g. Wang et al., 2022; Yang et al., 2016). Overall, these studies consistently indicate very low concentration levels in fish muscles, close to the concentrations in the water column and frequently below detection limits (e.g. MacMillan et al., 2017). Wang et al. (2022) specifically investigated the REE concentrations in the muscles of 14 species of marine fish sampled from an intensively fished area in China. Based on estimated daily intake levels, these authors then estimated a negligible risk for the local populations consuming these fish. Studies on the health risks associated with fish consumption seem destined to multiply due to the increase in anthropogenic releases of REEs into the environment. Furthermore, recent articles have investigated the organotropism of REEs in fish (e.g. Labassa et al., 2023; Lortholary et al., 2021; Marginson et al., 2023; Squadrone et al., 2020), or the subcellular distribution of REEs (e.g. Cardon et al., 2019, 2020) in aquatic organisms, reflecting the growing interest in understanding how natural or anthropogenic REEs are distributed and managed by higher organisms.

Another question in these studies is associated with questions about biomonitor species for emerging contaminants, which explains the abundance of data available for mollusks in the database (Fig. 2).

Indeed, the database comprises 43 % of concentration values in mollusks, with 51 % of these data concerning bivalve mollusks, which are now considered as excellent biomonitor species for TME contaminations, including REEs (e.g. [Bonnail et al., 2017](#); [Le Goff et al., 2019](#); [Merschel and Bau, 2015](#); [Pereto et al., 2020](#)). However, several authors have highlighted that other phyla, such as zooplankton ([Amyot et al., 2017](#); [MacMillan et al., 2019](#)), annelids ([Parisi et al., 2017](#)), sponges ([Orani et al., 2022](#)), or chlorophytes ([Gaudry et al., 2007](#)), can also serve as good biomonitor species for environmental monitoring of REEs.

3.1.3. Spatial distribution

Environmental contamination by REEs is currently a major issue in scientific research, both in freshwater and marine ecosystems. Out of the 102 articles in the database ([Fig. 3](#)), 75 provide concentration data for

marine organisms in uncontaminated environments, while 16 studies report cases of REE contamination in marine ecosystems. In freshwater ecosystems, 19 studies reported concentration levels in uncontaminated environments, compared to 11 studies in REE-contaminated environments.

The cases of REEs contamination could be classified into different categories: Some studies reported contamination in both organisms and the surrounding environment with several REEs, especially in the context of mining activities or mineral processing (e.g. [Blinova et al., 2021](#); [Bonnail et al., 2017](#); [Bosco-Santos et al., 2017](#); [Gaudry et al., 2007](#); [Palacios-Torres et al., 2020](#)). Other studies highlighted specific cases of contamination in organisms, particularly with Gd (e.g. [Akagi and Edanami, 2017](#); [Barrat et al., 2022](#); [Le Goff et al., 2019](#); [Pereto et al., 2020](#)) due to its use as a contrast agent in medical imaging ([Ebrahimi](#)

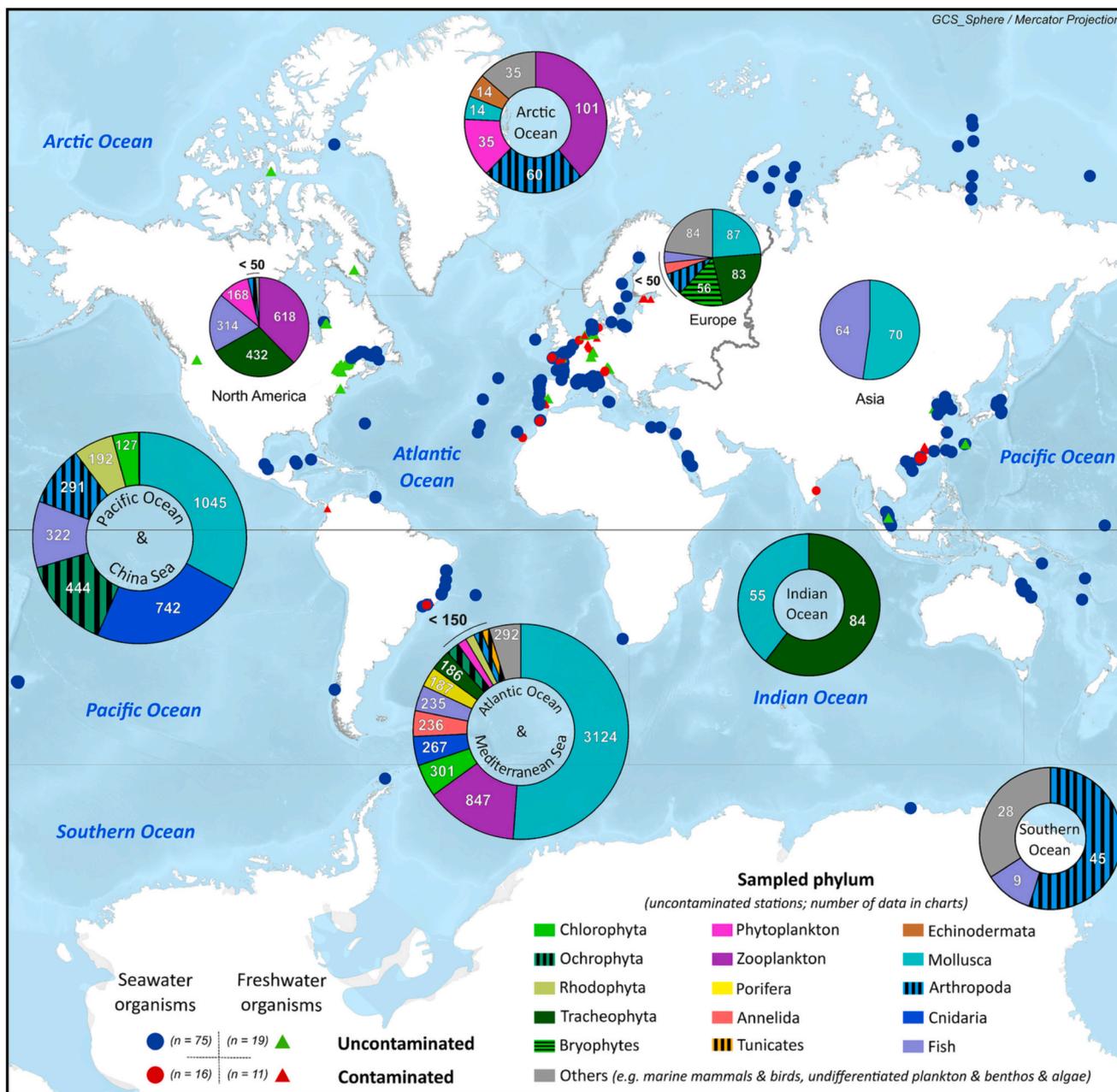


Fig. 3. Cartography of the 102 articles in the database ([Pereto et al., 2024](#)), based on the ecosystem type (circles or triangles) and the presence (red) or absence (blue and green) of REE contamination. The pie charts represent the number of available concentration data per phylum in uncontaminated freshwater ecosystems, according to geographic regions. The ring charts represent the number of available concentration data per phylum in uncontaminated marine ecosystems, according to geographic regions.

and Barbieri, 2019 and references therein). Finally, some studies highlighted the presence of anthropogenic REEs of industrial origin in water and aquatic organisms, with contamination in La and Ce (Bustamante and Miramand, 2005), La and Sm (Merschel and Bau, 2015), or Ce, Pr, and Nd (Ma et al., 2019).

Globally, most of the studies have been conducted near coastlines, particularly in Europe, China, and North America, in both freshwater and marine environments (Fig. 3). The majority of studies that reported cases of REE contamination have been conducted in Europe, with some cases in Asia and South America. The large amount of concentration data available in both freshwater and marine environments in North America, Europe, and Asia reflects, on one hand, the accessibility of scientific communities to ICP-MS analysis techniques, and on the other hand, the research topics previously mentioned. For example, in Canada, the announcement of potential REE mining sites (Amyot et al., 2017) lead scientific communities to establish natural concentration levels in regional ecosystems (e.g. Amyot et al., 2017; MacMillan et al., 2017). In Europe and Asia, the widespread use of REEs and their emergence as emerging contaminants in the environment as early as the 1990s (e.g. Bau and Dulski, 1996; Qiang et al., 1994) largely explain the abundance of studies in these regions.

In recent years, there has been an increasing observation of aquatic organisms contaminated with REEs (e.g. Barrat et al., 2022; Bosco-Santos et al., 2017; Merschel and Bau, 2015; Pereto et al., 2020; Wang et al., 2022), raising questions about reference levels of REE concentrations in aquatic ecosystems. Therefore, particular attention has been given to the natural REE concentrations in these organisms. The spatial distribution of data in uncontaminated environments has been studied across phyla in both freshwater (Fig. 3; pie charts) and marine (Fig. 3; rings) environments for different geographic regions. In freshwater environments, there are 2339 data available. The vast majority of the data was obtained in North America ($n = 1593$; zooplankton, tracheophytes, fishes) and Europe ($n = 366$; mollusks, tracheophytes, bryophytes). Some data are also available in Asia (mollusks, fishes). Most of the data comes from marine organisms ($n = 11,274$), particularly in the Atlantic Ocean and the Mediterranean Sea ($n = 6110$), followed by the Pacific Ocean and the China Sea ($n = 3167$). Both geographic regions are characterized by a large number of studied phyla and a significant number of data for mollusks, with $n = 3124$ and $n = 1045$, respectively. Finally, there are a few data points available for the Arctic Ocean ($n = 259$), the Indian Ocean ($n = 139$), and the Antarctic Ocean ($n = 82$), with zooplankton ($n = 101$), tracheophytes ($n = 84$), and arthropods ($n = 45$) as the predominant phyla, respectively. This mapping (Fig. 3) also reveals a number of data gaps. Globally, there is almost no information available for countries in Asia (except China and Japan), Africa, and South America (except Brazil). Moreover, very little information is currently available for freshwater ecosystems, particularly those far from the coast (e.g. major rivers, lakes). However, these continental environments are direct receptacles of many sources of REE contamination (e.g. Bonnail et al., 2017; Lerat-Hardy et al., 2021; Merschel and Bau, 2015; Pereto et al., 2023). These information gaps are particularly problematic for countries that exploit their REE resources. While several articles were published for China that address both health and environmental risks (e.g. Hao et al., 2015; Yang et al., 2016), other countries that exploit their REE resources are under-documented (e.g. USA, Australia). Mining activities, regardless of the element being extracted, are known for their significant impacts on aquatic ecosystems (e.g. Haque et al., 2014; Jain and Das, 2017). It is therefore crucial to fill these gaps, particularly through exploratory studies that define the levels of bioaccumulation within these pressured ecosystems.

Although this compilation of previously published articles over 50 years has certain limitations as mentioned earlier, these works lead to propose reference levels for the natural REE concentrations in several phyla, both in freshwater and marine ecosystems.

3.2. Reference concentrations of aquatic ecosystems

The correlation matrix has been conducted on the entire dataset of REE concentrations in aquatic organisms, and reveals strong positive and significant correlations among all REEs ($0.88 \leq r \leq 0.99$; p -value < 0.001 ; Supplementary Table A.2). Therefore, for simplifying further analyses, we focused on three elements that represent the three groups of REEs: i) La for LREEs; ii) Gd for MREEs; and iii) Yb for HREEs. Gadolinium was chosen here because we have estimated that the increasing number of articles reporting cases of environmental contamination with Gd (e.g. Akagi and Edanami, 2017; Bau and Dulski, 1996; Castro et al., 2023; Pereto et al., 2023) necessitates the establishment of reference concentrations for the scientific community.

3.2.1. Abiotic compartments

The database includes concentrations of REEs in waters ($n = 535$) and sediments ($n = 1125$) from uncontaminated freshwater and marine ecosystems. The data were compared to a few large-scale studies in order to assess if these concentration ranges represent global natural levels.

For freshwater ecosystems, there are limited sediment concentration values available in the database, with 61 data points but no value for Eu, Tb, Ho, Tm, and Lu. Viers et al. (2009) estimated REE concentrations in suspended particulate matter from global rivers, with a total mean concentration ($\sum \text{REE}_{\text{mean}}$) of $174,830 \text{ ng.g}^{-1}$. On the European scale, data from the FOREGS program (Forum of European Geological Surveys; Salminen et al., 2005) defined a $\sum \text{REE}_{\text{mean}}$ of $198,900 \text{ ng.g}^{-1}$ and a median total concentration ($\sum \text{REE}_{\text{med}}$) of $158,400 \text{ ng.g}^{-1}$ for river sediments. Regarding freshwater, the FOREGS program also provided REEs concentration data for European river waters, with a $\sum \text{REE}_{\text{med}}$ of 188 ng.L^{-1} , which is consistent with the concentrations in the database ($n = 147$; $138 \pm 174 \text{ ng.L}^{-1}$; Table 1). The partition coefficient (Kd) between the two compartments is approximately $1,100,000 \text{ L.kg}^{-1}$ ($\log Kd = 6.1 \text{ L.kg}^{-1}$).

For marine ecosystems, the 591 data points compiled by Piarulli et al. (2021) were compared to the 1064 sediment data points available in the database (Table 1, Supplementary Fig. A.1). Regardless of the specific REE considered, the REE concentrations in the database sediments ($\sum \text{REE}_{\text{med}} = 120,400 \pm 131,000 \text{ ng.g}^{-1}$) were not significantly different from the concentrations in coastal or oceanic marine sediments (p -value > 0.05). Regarding seawater (Table 1), the 336 data points from Kuss et al. (2001), as well as the 592 data points compiled by Neira et al. (2022) and the 404 data points compiled by Piarulli et al. (2021), were compared to the 388 concentration values in the database (Supplementary Fig. A.1). Due to a large number of studies conducted in coastal environments, the seawater concentrations in the database ($\sum \text{REE}_{\text{med}} = 36.1 \pm 35.0 \text{ ng.L}^{-1}$; Table 1) are similar to the concentrations in coastal environments reported in the literature (p -value > 0.05) and significantly higher than the concentrations in open oceans (p -value < 0.05). The partition coefficient between the two abiotic compartments is approximately $3,300,000 \text{ L.kg}^{-1}$ ($\log Kd = 6.5 \text{ L.kg}^{-1}$).

The REE concentrations in the abiotic compartments are separated by six orders of magnitude, both in freshwater and marine ecosystems. This observation confirms the lithophilic nature of REEs and their strong affinity for the particulate fraction on a large scale (Brookins, 1989; Gaillardet et al., 2003; Merschel et al., 2017). The partition coefficient calculated for marine ecosystems is larger than that of freshwater ecosystems. This difference is mainly due to the reactivity of REEs along the land-ocean continuum and within the oceans themselves (e.g. Elderfield and Greaves, 1982; Lawrence and Kamber, 2006), leading to very low concentrations in seawater. At the land-ocean interface, dissolved REEs are known for their reactivity to estuarine salinity gradients (Elderfield et al., 1990; Sholkovitz and Szymczak, 2000). In rivers, REEs are largely associated with stable, iron-rich organic colloids (Elderfield et al., 1990; Sholkovitz, 1995). At the initial salinity points ($S < 5$), the coagulation-flocculation of organic colloids and gravity sedimentation, leads to a significant decrease in REE concentrations in the dissolved fraction

Table 1
Median concentrations of REEs ($n \geq 5$; \pm MAD) and sum of median REE concentrations (\sum REEs_{med}; \pm \sum MAD) at a global scale, for abiotic compartments (freshwater and seawater in $\mu\text{g.L}^{-1}$; sediments in ng.g^{-1}) and phyla for freshwater and marine ecosystems (ng.g^{-1}). Number of associated data (n).

Freshwater ecosystems	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	\sum REEs _{med} \pm \sum MAD
Freshwater	0.0298 \pm 0.0405 (12)	0.0381 \pm 0.0519 (11)	0.0055 \pm 0.0069 (11)	0.03 \pm 0.0373 (11)	0.0086 \pm 0.011 (11)	0.0021 \pm 0.0027 (11)	0.0067 \pm 0.0066 (11)	0.0012 \pm 0.0014 (10)	0.006 \pm 0.006 (11)	0.0013 \pm 0.0014 (10)	0.0036 \pm 0.0039 (10)	0.0007 \pm 0.0009 (10)	0.0027 \pm 0.0023 (8)	0.0013 \pm 0.0016 (10)	0.138 \pm 0.174
Phytoplankton	27400 \pm 3450 (12)	56900 \pm 7020 (12)	7500 \pm 1120 (12)	29200 \pm 4810 (12)	5540 \pm 837 (12)	673 \pm 97.2 (12)	4690 \pm 723 (12)	607 \pm 97.5 (12)	3390 \pm 574 (12)	650 \pm 114 (12)	1760 \pm 321 (12)	211 \pm 35.8 (12)	1450 \pm 257 (12)	206 \pm 38.9 (12)	140100 \pm 19500
Zooplankton	3210 \pm 4490 (41)	4960 \pm 6830 (41)	759 \pm 1060 (41)	2590 \pm 3600 (41)	486 \pm 675 (41)	90.1 \pm 117 (39)	414 \pm 575 (41)	54.8 \pm 76.3 (41)	309 \pm 430 (41)	59.7 \pm 83.2 (41)	165 \pm 231 (41)	21.2 \pm 29.9 (41)	144 \pm 202 (41)	21.3 \pm 29.7 (41)	13300 \pm 18400
Molusca	1120 \pm 725 (8)	2660 \pm 2480 (8)	261 \pm 191 (8)	990 \pm 683 (8)	187 \pm 129 (8)	29.4 \pm 13 (7)	551 \pm 558 (8)	34.9 \pm 23 (7)	158 \pm 95.3 (8)	28 \pm 14.7 (7)	96 \pm 46 (7)	11.9 \pm 2.34 (7)	81.8 \pm 37.3 (7)	13.7 \pm 1.4 (7)	6220 \pm 5000
Tracheophyta	905 \pm 1190 (11)	2040 \pm 2680 (11)	202 \pm 254 (11)	824 \pm 1060 (11)	182 \pm 241 (11)	16.3 \pm 13.3 (5)	208 \pm 280 (11)	8.23 \pm 6.58 (6)	159 \pm 216 (11)	8.97 \pm 6.95 (6)	85.2 \pm 117 (11)	3.3 \pm 2.54 (6)	70 \pm 96.8 (11)	2.93 \pm 2.05 (6)	4710 \pm 6170
Fish	25.5 \pm 29.7 (20)	40.5 \pm 41.5 (20)	5.5 \pm 5.93 (20)	21.5 \pm 23 (20)	9 \pm 10.4 (13)	4 \pm 2.97 (13)	9 \pm 8.9 (13)	10 \pm 2.97 (7)	4 \pm 2.97 (15)	1 \pm 1 (9)	2.5 \pm 1.48 (12)		9 \pm 7.41 (7)		136 \pm 137
Marine ecosystems	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	\sum REEs _{med} \pm \sum MAD
Sediment	23600 \pm 26200 (82)	55300 \pm 59700 (81)	5890 \pm 6300 (76)	20900 \pm 22800 (78)	3270 \pm 3450 (79)	900 \pm 1020 (79)	3150 \pm 3200 (76)	575 \pm 669 (78)	2810 \pm 3190 (68)	670 \pm 801 (75)	1520 \pm 1700 (68)	250 \pm 297 (75)	1320 \pm 1380 (71)	230 \pm 282 (78)	120400 \pm 131000
Seawater	0.008 \pm 0.0087 (29)	0.0117 \pm 0.0129 (29)	0.0015 \pm 0.0014 (28)	0.0058 \pm 0.0056 (29)	0.0013 \pm 0.0011 (29)	0.0003 \pm 0.0003 (29)	0.0018 \pm 0.0013 (29)	0.0003 \pm 0.0002 (24)	0.0018 \pm 0.001 (29)	0.0005 \pm 0.0002 (28)	0.0014 \pm 0.0009 (29)	0.0002 \pm 0.0001 (23)	0.0013 \pm 0.001 (29)	0.0002 \pm 0.0002 (24)	0.0361 \pm 0.035
Tunicates	1780 \pm 758 (5)	4110 \pm 2090 (5)	478 \pm 246 (5)	1980 \pm 940 (5)	487 \pm 274 (5)	111 \pm 47.4 (5)	474 \pm 222 (5)	60 \pm 29.7 (5)	279 \pm 138 (5)	47 \pm 25.2 (5)	122 \pm 72.6 (5)	12 \pm 4.45 (5)	71 \pm 32.6 (5)	8 \pm 2.97 (5)	10000 \pm 4890
Porifera	1250 \pm 615 (20)	1100 \pm 801 (20)	130 \pm 141 (9)	700 \pm 771 (11)	135 \pm 148 (12)	24 \pm 31.1 (9)	130 \pm 59.3 (21)	10 \pm 10.4 (8)	110 \pm 51.9 (20)	18 \pm 22.2 (9)	60 \pm 36.3 (16)	2 \pm 1.48 (7)	50.5 \pm 19.3 (18)	2 \pm 1.48 (7)	3720 \pm 2710
Chlorophyta	1460 \pm 1560 (32)	1290 \pm 1230 (35)	230 \pm 193 (29)	1080 \pm 1140 (32)	215 \pm 209 (32)	49.8 \pm 42.6 (35)	115 \pm 153 (15)	36.5 \pm 41.6 (30)	210 \pm 192 (29)	48 \pm 45.1 (28)	130 \pm 121 (28)	17 \pm 14.8 (28)	92 \pm 83.8 (34)	16 \pm 14.8 (33)	4980 \pm 5050
Ochrophyta	540 \pm 689 (47)	1750 \pm 2380 (56)	420 \pm 371 (29)	1110 \pm 1510 (46)	120 \pm 160 (45)	60 \pm 46 (41)	360 \pm 326 (29)	50 \pm 39.5 (36)	260 \pm 192 (29)	50 \pm 31.1 (28)	140 \pm 89 (29)	20 \pm 10.4 (28)	92 \pm 91.9 (50)	16.5 \pm 9.71 (36)	4990 \pm 5950
Rhodophyta	520 \pm 25 (25)	1880 \pm 2300 (28)	111 \pm 146 (14)	350 \pm 452 (19)	75.6 \pm 96.2 (24)	45.2 \pm 55.1 (24)	35.9 \pm 32.9 (14)	18.3 \pm 24.2 (22)	25.8 \pm 22.2 (14)	6.81 \pm 5.19 (13)	15.7 \pm 14.2 (13)	2.09 \pm 1.51 (13)	47.7 \pm 61.6 (26)	10.5 \pm 13.9 (22)	3140 \pm 3950
Molusca	321 \pm 111 (11)	507 \pm 446 (11)	74.1 \pm 66.9 (105)	295 \pm 255 (107)	67 \pm 57.8 (105)	11.5 \pm 9.68 (103)	60.5 \pm 49.8 (105)	10 \pm 7.71 (99)	49 \pm 41.5 (103)	10 \pm 8.9 (105)	29 \pm 26.7 (105)	3.1 \pm 3.11 (91)	21 \pm 20.4 (105)	3 \pm 2.97 (90)	1460 \pm 1300
Arthropoda	30 \pm 29.8 (74)	69.5 \pm 81.7 (70)	26.5 \pm 20 (12)	54 \pm 72.6 (35)	12.7 \pm 17.3 (38)	5 \pm 4.45 (11)	16 \pm 13.3 (11)	3.88 \pm 4.63 (8)	14 \pm 13.6 (11)	4.23 \pm 4.88 (7)	7 \pm 6.97 (11)	1.8 \pm 1.47 (7)	5 \pm 6.89 (38)	1.77 \pm 1.42 (7)	251 \pm 279
Cnidaria	13 \pm 11.9 (78)	16.4 \pm 10.2 (78)	2.85 \pm 2.22 (68)	10.6 \pm 8.01 (78)	2.54 \pm 1.81 (78)	1.16 \pm 1.25 (57)	4 \pm 1.85 (75)	0.56 \pm 0.29 (66)	3.4 \pm 2.37 (75)	0.958 \pm 0.587 (68)	2.55 \pm 2.29 (78)	0.411 \pm 0.369 (66)	2.67 \pm 2.58 (78)	0.4 \pm 0.393 (66)	61.5 \pm 46.1
Fish	15.1 \pm 20.9 (11)	24 \pm 33.2 (11)	7.57 \pm 10.6 (11)	7.3 \pm 9.41 (11)	3.16 \pm 4.2 (10)	0.47 \pm 0.482 (6)	1.32 \pm 0.845 (9)	1 \pm 1.36 (7)	2.22 \pm 2.56 (11)	0.512 \pm 0.59 (6)	0.64 \pm 0.786 (9)	0.186 \pm 0.231 (6)	2.18 \pm 2.97 (9)	0.05 \pm 0.0445 (5)	65.7 \pm 88.2
Phytoplankton	4.02 \pm 3.84 (9)	5.74 \pm 4.14 (9)	0.723 \pm 0.5 (9)	3.18 \pm 2.25 (9)	0.775 \pm 0.559 (9)	0.178 \pm 0.135 (9)	0.794 \pm 0.58 (9)	0.079 \pm 0.04 (7)	0.588 \pm 0.439 (9)	0.103 \pm 0.0786 (9)	0.242 \pm 0.153 (8)	0.025 \pm 0.0163 (7)	0.223 \pm 0.172 (9)	0.021 \pm 0.0148 (7)	16.7 \pm 12.9
Zooplankton	29.4 \pm 43.1 (69)	42.2 \pm 61.8 (69)	5.39 \pm 7.91 (69)	19.5 \pm 28.6 (69)	3.27 \pm 4.77 (66)	0.398 \pm 0.578 (60)	3.09 \pm 4.49 (66)	0.347 \pm 0.502 (69)	2.05 \pm 2.97 (69)	0.391 \pm 0.568 (69)	1 \pm 1.45 (69)	0.143 \pm 0.208 (66)	0.8 \pm 1.16 (69)	0.129 \pm 0.189 (68)	108 \pm 158

REEs concentrations in aquatic environments (median \pm MAD; ng.g^{-1} ; $\mu\text{g.L}^{-1}$ for water) and number of data (n).

(Lawrence and Kamber, 2006 and references therein). This phenomenon is not uniform along the lanthanide series, resulting in fractionations. Indeed, LREEs seem more affected than MREEs, which are, in turn, more affected than HREEs (Lawrence and Kamber, 2006; Nozaki et al., 2000). These differences in reactivity are linked to the stability of REEs in the dissolved fraction because the lanthanide contraction implies higher stability of dissolved complexes with increasing atomic number (Elderfield et al., 1988). The estuarine processes partially explain the concentration differences observed between freshwater and coastal seawater.

Overall, the concentrations obtained for the abiotic compartments in the database are representative of natural concentrations on a global scale. These concentrations can be used subsequently to study the reference levels of REE concentrations in the context of global biogeochemical cycles.

3.2.2. REE concentrations in aquatic organisms

The contamination of aquatic ecosystems through anthropogenic releases of REEs is at the forefront of scientific investigations surrounding the study of REE bioaccumulation. The challenges which face the emerging contaminants require to understand the natural levels of REE concentrations in the ecosystems, particularly in aquatic organisms. Reference levels of REE concentrations are essential tools for identifying and quantifying the exposure levels of ecosystems to these emerging contaminants. While knowledge regarding the geochemical behavior of REEs has long been available (e.g. Lawrence and Kamber, 2006; Migaszewski and Galuszka, 2015 and references therein), information concerning their concentration in organisms is still fragmented (e.g. Neira et al., 2022; Piarulli et al., 2021; Rétif et al., 2023). Based on this compilation of studies, we focused on data regarding whole organisms in uncontaminated environments to establish reference levels of REE concentrations.

For freshwater ecosystems, only a few articles provide concentration data on whole organisms ($n \geq 5$) in uncontaminated environments. An assessment of concentration levels can be proposed for only 5 phyla (Table 1): phytoplankton, zooplankton, mollusks, tracheophytes, and fishes. For marine ecosystems, reference concentration levels can be defined for 11 phyla (Table 1): tunicates, ochrophytes, chlorophytes, sponges, rhodophytes, mollusks, arthropods, zooplankton, cnidarians, fishes, and phytoplankton. Only the data concerning cnidarians (i.e. hard corals in the database) are not whole-organism data but rather concentrations for their calcium skeletons. Using MAD (Median Absolute Deviation), significant dispersions can be observed among the different phyla (Table 1). The complete distribution of available concentration data for whole organisms has been studied for each REE (Supplementary Fig. A.2), including the three selected REEs (La, Gd, Yb; Fig. 4). These distributions have been defined for marine organisms (Fig. 4A) and freshwater organisms (Fig. 4B) and highlight several key findings.

In this study, four phyla are common to both freshwater and marine ecosystems. Regardless of the phylum, REE concentrations are higher in freshwater organisms, particularly in planktonic species (i.e. phytoplankton, zooplankton). This supports the natural variations in concentrations observed between freshwater, which is more concentrated in REEs, and seawater, which is less concentrated (e.g. Elderfield et al., 1990; Lawrence and Kamber, 2006; Sholkovitz, 1995). Moreover, the differences in concentration between freshwater and marine organisms can be related to changes in the speciation and bearing-phases of REEs between both ecosystems (Smrzka et al., 2019 and references therein). It appears that planktonic species are particularly influenced by these differences, suggesting a significant influence of environmental concentrations on REE concentrations in organisms. Previous studies highlighted that concentrations of REEs in the form of free ions (REE^{3+}) are good predictors of concentrations measured in plankton (MacMillan et al., 2019; Strady et al., 2015). Changes in speciation at the continent-ocean interface can partly explain the significant differences observed

between freshwater and marine ecosystems for phytoplankton and zooplankton. Similar observations have also been made by Dang et al. (2023), Marginson et al. (2023) and Lobus et al. (2019).

In freshwater ecosystems (Fig. 4B; Supplementary Fig. A.2), phytoplankton consistently has the highest REE concentrations, followed by zooplankton. In contrast, fishes exhibit concentration levels that are not significantly different ($p\text{-value} > 0.05$) from the surrounding water, regardless of the REE. Thus, there is a factor of 1000 separating the highest levels (i.e. phytoplankton) from the lowest REE concentration levels (i.e. fish). On the other hand, mollusks and tracheophytes have intermediate concentrations between zooplankton and fishes. These phyla are distributed in the following decreasing order: phytoplankton > zooplankton > mollusks > tracheophytes > fishes > freshwater. It is important to note that only one study reported REE concentrations in phytoplankton (Dang et al., 2023) and fish (whole organisms; Mayfield and Fairbrother, 2015), and only two studies exist for tracheophytes (Cowgill, 1973; Zocher et al., 2022). Therefore, understanding the distribution of REE concentrations in freshwater ecosystems remains complex. Due to the limited information available for freshwater ecosystems, the subsequent focus of this study will be on marine ecosystems.

In marine ecosystems, there is a factor of 600 separating the highest levels (i.e. tunicates) from the lowest REE concentration levels (i.e. phytoplankton). Within these ecosystems, three groups of phyla can be identified between both abiotic compartments (Fig. 4A; Supplementary Fig. A.2): i) a group of phyla for which the concentrations of at least one REE are not different from those in sediments (i.e. tunicates, sponges, ochrophytes, chlorophytes); ii) a group of phyla where the concentrations of all 14 REEs are significantly different from sediment and seawater concentrations (i.e. rhodophytes, mollusks); and iii) a group of phyla for which the concentrations of at least one REE are not different from seawater concentrations (i.e. arthropods, cnidarians, zooplankton, fish, phytoplankton). This large-scale phylum-based approach seems to reflect the combined influences of habitat and trophic position on observed concentrations.

3.2.3. Influences of habitat and trophic position

The potential impact of anthropogenic REEs on aquatic ecosystems requires a better understanding of the REE dynamics within food webs and the factors controlling these dynamics. Direct and trophic transfer pathways, along with detoxification capacities, determine the concentration levels of organisms for both TMEs and REEs. These transfer pathways are heavily influenced by the bioavailability of REEs in the environment and in the food sources of aquatic organisms.

In marine ecosystems (Fig. 4A; Supplementary Fig. A.2), regardless of the studied REE, all organisms exhibit REE concentration levels that are comprised between the two abiotic compartments (sediments > organisms > water). Furthermore, the partition coefficient ($K_d = 3,300,000 \text{ L.kg}^{-1}$) that separates the concentrations of the two abiotic compartments can have a significant effect on the observed concentration levels, depending on whether the organism is associated with one or the other compartment (e.g. benthic or pelagic species). Three groups of phyla have been observed, suggesting influences from habitat (i.e. living environment) and/or trophic position (i.e. food sources) of the species. The first group (i.e. tunicates, sponges, ochrophytes, chlorophytes), which is close to the concentrations in the sediment, is exclusively composed of benthic species that are fixed to the substrate and have a low trophic level. In contrast, the third group consists of pelagic species (e.g. phytoplankton, zooplankton) and/or species at higher trophic levels (e.g. fishes). Only the cnidarians seem to deviate from this trend as the available data correspond to the low REE concentrations in their calcium carbonate skeletons.

The influence of habitat can be demonstrated in marine organisms (Fig. 5A, Supplementary Fig. A.3), where REE concentrations can be ranked as follows: sediments > benthic species > pelagic species > seawater. Therefore, benthic organisms have significantly higher REE

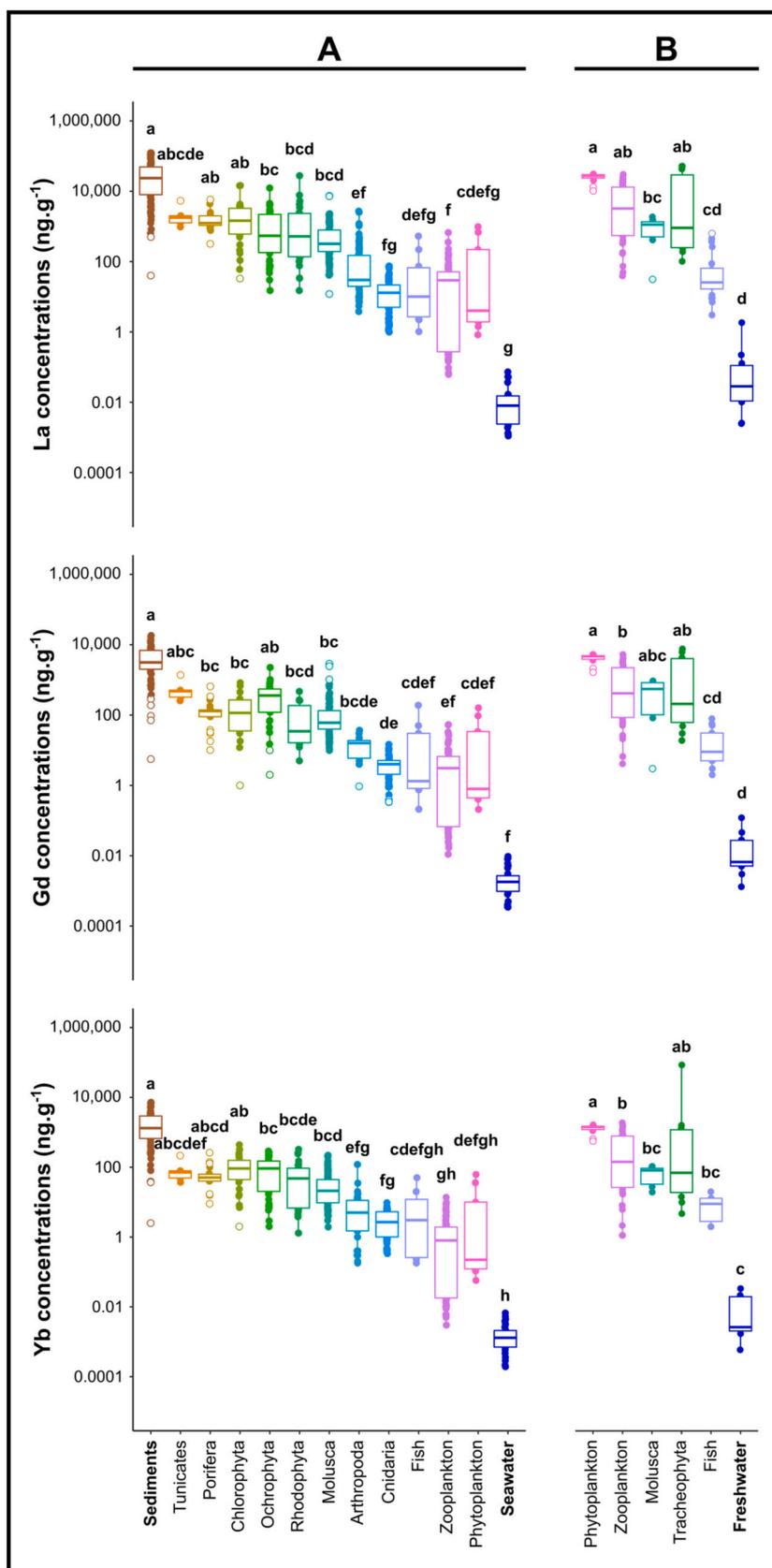


Fig. 4. Concentrations of La, Gd, and Yb ($\text{ng}\cdot\text{g}^{-1}$; $\mu\text{g}\cdot\text{L}^{-1}$ for water) in abiotic and biotic compartments of marine ecosystems (A) and freshwater ecosystems (B) according to organism phylum. Non-parametric Kruskal-Wallis test ($\alpha = 0.05$), Dunn's post-hoc test ($\alpha = 0.05$), and Bonferroni correction were applied. Outliers are represented by empty circles. The distribution of all REEs is available in Supplementary Fig. A.2.

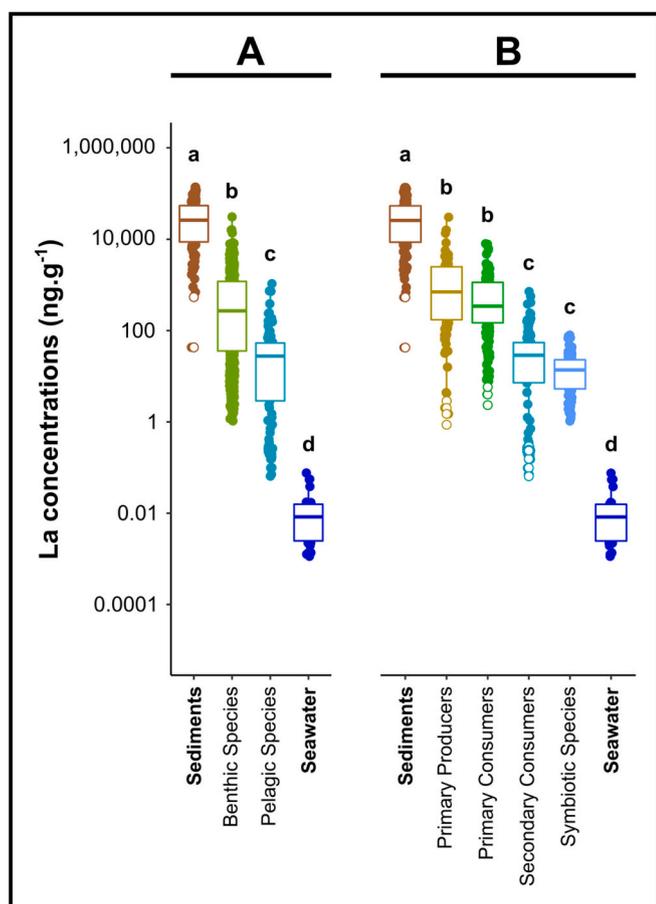


Fig. 5. Concentrations of La (ng.g^{-1} ; $\mu\text{g.L}^{-1}$ for water) in abiotic and biotic compartments of marine ecosystems according to: A) habitat (i.e. benthic species, pelagic species); and B) trophic position (i.e. primary producers, primary and secondary consumers, symbiotic species). Non-parametric Kruskal-Wallis test ($\alpha = 0.05$), Dunn's post-hoc test ($\alpha = 0.05$), and Bonferroni correction were applied. Outliers are represented by empty circles. The distribution of all REEs is available in Supplementary Fig. A.3.

concentration levels ($p\text{-value} < 0.05$) compared to pelagic organisms. In contrast, pelagic organisms exhibit REE concentrations that are similar to those in the water column. These observations are consistent with previous studies (e.g. MacMillan et al., 2017; Rétif et al., 2024; Wang et al., 2022; Yang et al., 2016) conducted in aquatic environments but at smaller spatial scales. These results demonstrate that the habitat of marine organisms appears to have a major influence on REE concentrations at the global scale.

If we considered the REE concentrations based on the trophic positions of marine organisms (Fig. 5B, Supplementary Fig. A.3), the following ranking can be observed: primary producers = primary consumers > secondary consumers = symbiotic species. Organisms at lower trophic levels has the highest REE concentration levels. These levels decrease with increasing trophic position of the organisms, indicating a trophic dilution of REE concentrations. The trophic position appears to be a controlling factor for the REE concentrations in marine organisms. These observations are consistent with previous regional studies on freshwater (Amyot et al., 2017; Weltje et al., 2002) and marine (MacMillan et al., 2017; Squadroni et al., 2019; Wang et al., 2019) ecosystems. Mayfield and Fairbrother (2015) measured significantly higher concentrations of REEs in benthivorous fish compared to piscivorous fish. Amyot et al. (2017) demonstrated similar results with non-predatory benthic invertebrates with higher concentration levels than predatory benthic invertebrates. In the marine environment, Battuello et al. (2017) observed a decrease in concentration levels of La and Ce

among herbivorous, omnivorous, and carnivorous zooplankton species. More recently, other authors were also able to demonstrate trophic dilution of REE concentrations along estuarine food webs (Rétif et al., 2024; Santos et al., 2023). In conclusion, the collective observations highlighted that the REE concentration levels in aquatic organisms are strongly influenced by their habitat and position within food webs. Globally, there is a trophic dilution of natural REE concentrations and an absence of biomagnification along food webs. These large scale observations provide insights into the dynamics of REEs within food webs and contribute to an overall understanding of global biogeochemical cycles of REEs.

3.3. Research perspectives

Several significant findings have emerged from this study and highlighted a number of gaps in our understanding of REE concentrations in aquatic organisms. Differences in data availability among REEs (i.e. lack of data for HREEs), geographic regions (e.g. Indian Ocean, Central Europe, Africa), and taxonomic groups have resulted in imbalances in the database used in this study. These observations provide insights into research strategies that should be prioritized in the coming years.

From a spatial perspective, it has been observed that certain geographic areas are under-represented at a global scale. Conversely, regions such as the coasts of the Atlantic Ocean and the Mediterranean Sea appear to be well-documented. However, when considering the environments studied, it is evident that critical zones between land and ocean (e.g. estuaries, deltas, lagoons, mangroves) remain poorly studied. These areas are known for their biodiversity and the numerous ecosystem services they provide (e.g. Barbier et al., 2011; Ligorini et al., 2023; Twilley et al., 2017). These ecosystems are sensitive, characterized by fragile balances, and under significant anthropogenic pressures (e.g. García-Pintado et al., 2007; Teichert et al., 2018; Zucchetta et al., 2021), including REE inputs (e.g. Bosco-Santos et al., 2017; Brito et al., 2018; Elbaz-Poulichet et al., 2002; Pereto et al., 2023). Therefore, further studies require to characterize the REE concentrations in organisms within these vulnerable ecosystems. The establishment of observation networks, encompassing both abiotic compartments (e.g. water, suspended matter, sediments) and aquatic organisms, appears to be an appropriate solution for this purpose (Briant et al., 2021; Ma et al., 2019; Pereto et al., 2023).

At a global scale, while the mollusks are well-represented (i.e. 43 % of the database), key phyla in ecosystems such as annelids, echinoderms, bryophytes, and phytoplankton are poorly represented. These gaps are particularly pronounced in freshwater ecosystems. For example, the limited available data for arthropods hinders the assessment of reference concentration levels for this phylum. Furthermore, certain groups of species, such as phytobenthos, are completely absent, despite their major ecological role in freshwater ecosystems. These ecosystems are directly influenced by natural inputs related to the mechanical erosion and the leaching of geological outcrops in the watersheds and are therefore subject to greater geographical variability driven by natural geochemical background (e.g. Gaillardet et al., 2003; Sholkovitz, 1995; Viers et al., 2009). Additionally, these environments receive increasing inputs of anthropogenic REEs (e.g. Kulaksız and Bau, 2013; Lerat-Hardy et al., 2019). It is therefore necessary to characterize the abiotic compartments when studying REE concentrations in aquatic organisms. Furthermore, it is crucial to deepen our knowledge of the natural concentrations in under-represented phyla to better understand the dynamics of REEs. This study aims at providing a database that can serve as a support for the scientific communities to fill these gaps. These new insights will ultimately allow for the identification and characterization of the disturbances caused by anthropogenic REEs, while considering the natural geochemical background in these environments.

Due to the emergence of environmental contamination by REEs in recent years, studies demonstrating bioaccumulation of anthropogenic

REEs are increasingly appearing in the literature but remain under-documented. While several studies have shown the presence of REE contamination in aquatic organisms, only a few phyla are concerned, primarily bivalve mollusks (e.g. Akagi and Edanami, 2017; Barrat et al., 2022; Bonnail et al., 2017; Le Goff et al., 2019; Merschel and Bau, 2015; Pereto et al., 2020). However, a few studies highlighted contamination cases in chlorophytes (Gaudry et al., 2007), arthropods (Bosco-Santos et al., 2017), and fishes (Wang et al., 2022). Thus, current knowledge on the dynamics of anthropogenic REEs is still fragmented and does not allow for the assessment of the transfer of these contaminants within food webs. The available knowledge on inter-organ transfers of these contaminants is also incomplete. It is therefore essential to address these gaps in order to develop this knowledge further and better characterize the dynamics of these emerging contaminants, and possibly predict their future evolutions.

Finally, the combined influences of habitat and trophic position of organisms appear to be major factors controlling the concentration of REEs. However, it remains challenging to decipher the effects of habitat from those of trophic position. In geochemistry, the study of REEs, particularly their behavior, is predominantly based on the analysis of normalized patterns (Rétif et al., 2023 and references therein). These patterns are obtained by dividing the REE concentrations of the sample by those of a geochemical reference material such as PAAS (Post-Archean Australian Shale; Nance and Taylor, 1976; Pourmand et al., 2012), NASC (North American Shale Composite; McLennan, 1989), or Chondrite (Anders and Grevesse, 1989; Barrat et al., 2012). The study of these patterns and their characteristics (i.e. fractionations) allows for an understanding of environmental processes and visual identification of anthropogenic REEs (i.e. distinguishing natural and anthropogenic contributions). Many studies reported normalized patterns to identify REE fractionations and understand the dynamics of these elements (e.g. Akagi and Edanami, 2017; Figueiredo et al., 2022b; Merschel and Bau, 2015; Zocher et al., 2022). For example, the combined study of patterns from water (Rhine River, Germany) and shells (bivalve mollusk *Corbicula fluminea*) allowed to highlight the transfer of anthropogenic La and Sm from water to shells and the absence of transfer of anthropogenic Gd of medical origin present in the water (Merschel and Bau, 2015). Other studies linked the signatures of natural REEs observed in the watershed or at specific stations with those observed in aquatic organisms (e.g. Pastorino et al., 2020; Wang et al., 2022; Weltje et al., 2002). The use of normalized patterns as fingerprints appears to be a relevant approach to attempt to separate the influences of habitat and trophic position. These research perspectives require the use of suitable normalizing reference materials for studying REE bioaccumulation. However, currently, no biological normalizing reference materials exists (Rétif et al., 2023). Furthermore, the use of local normalizing materials to study normalized patterns of aquatic organisms (i.e. water, sediment, prey of the studied organisms) can prove to be promising perspectives for studying the transfers of REEs at the interface between the environment and organisms and within trophic food webs.

The originality of this compilation of studies is to provide, for the first time, a global approach to the currently available data and establish ranges of REE concentrations in aquatic organisms. These concentration ranges can then serve as reference levels for the natural REE concentrations on a large scale. These references can support the scientific community in better contextualizing their work on a global scale and enable the characterization of potential disturbances caused by anthropogenic REEs. Nevertheless, it is important to emphasize that this study is a first step towards a global understanding of the distribution of REEs in aquatic organisms. It is for these reasons that the database used is accessible to the scientific community, so that it can be reused, supplemented and reinterpreted through new issues by recent literature published since February 2023 (e.g. Marginson et al., 2023; Pastorino et al., 2024; Rétif et al., 2024; Santos et al., 2023).

CRediT authorship contribution statement

Clément Pereto: Conceptualization, Data curation, Formal analysis, Investigation, Visualization, Writing – original draft. **Magalie Baudrimont:** Supervision, Writing – review & editing. **Alexandra Coynel:** Funding acquisition, Project administration, Resources, Supervision, Validation, Writing – review & editing.

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.

Data availability

All the data used is available online on PANGAEA. The reference for this database is specified in the manuscript.

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Appendix A. Supplementary data

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