Environmental Pollution 291 (2021) 118230

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

# Sources, distribution and effects of rare earth elements in the marine environment: Current knowledge and research gaps☆

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## ARTICLE INFO

*Keywords:*
REY
Marine ecosystem
Distribution
Transfer
Ecotoxicity
Risk assessment

## ABSTRACT

Rare earth elements and yttrium (REY) are critical elements for a wide range of applications and consumer products. Their growing extraction and use can potentially lead to REY and anthropogenic-REY chemical complexes (ACC-REY) being released in the marine environment, causing concern regarding their potential effects on organisms and ecosystems. Here, we critically review the scientific knowledge on REY sources (geogenic and anthropogenic), factors affecting REY distribution and transfer in the marine environment, as well as accumulation in- and effects on marine biota. Further, we aim to draw the attention to research gaps that warrant further scientific attention to assess the potential risk posed by anthropogenic REY release. Geochemical processes affecting REY mobilisation from natural sources and factors affecting their distribution and transfer across marine compartments are well established, featuring a high variability dependent on local conditions. There is, however, a research gap with respect to evaluating the environmental distribution and fate of REY from anthropogenic sources, particularly regarding ACC-REY, which can have a high persistence in seawater. In addition, data on organismal uptake, accumulation, organ distribution and effects are scarce and at best fragmentary. Particularly, the effects of ACC-REY at organismal and community levels are, so far, not sufficiently studied.

To assess the potential risks caused by anthropogenic REY release there is an urgent need to i) harmonise data reporting to promote comparability across studies and environmental matrices, ii) conduct research on transport, fate and behaviour of ACC-REY *vs* geogenic REY iii) deepen the knowledge on bioavailability, accumulation and effects of ACC-REY and REY mixtures at organismal and community level, which is essential for risk assessment of anthropogenic REY in marine ecosystems.

## 1. Introduction

Rare earth elements, comprising the 15 lanthanoides and yttrium (hereafter REY) feature relatively uniform physical-chemical properties and are, despite their name, widely distributed in the earth's crust (USEPA, 2012). Rare earth elements and yttrium usually co-occur as either minor or major constituents in ores/accessory minerals (Balaram, 2019; Wang et al., 2019). REY can be divided into different classes according to their electron configuration. Most commonly they are classified into light REY (LREY), comprising lanthanum (La), cerium (Ce), praseodymium (Pr), neodymium (Nd) samarium (Sm) and europium (Eu), and heavy REY (HREY) that include gadolinium (Gd), terbium (Tb), dysprosium (Dy), holmium (Ho), erbium (Er), thulium (Tm), ytterbium (Yb), lutetium (Lu) and yttrium (Y) (Gonzalez et al., 2014). In

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☆ This paper has been recommended for acceptance by Wen-Xiong Wang.

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https://doi.org/10.1016/j.envpol.2021.118230
Received 19 August 2021; Received in revised form 21 September 2021; Accepted 23 September 2021

several cases, REY are additionally grouped into medium REY (MREY), most commonly encompassing Sm, Eu, Gd, Tb, Dy, Ho and Y. However, in this review we will hereafter mostly refer to light and heavy REY, as the grouping of MREY is not always consistent in literature.

REY electron structure is characterised by electrons in the 4f orbital that are shielded by the 5s and 5p electrons, which gives them characteristics catalytic, magnetic and electronic properties (e.g., high density and melting point and high conductivity). This makes REY essential components for a wide range of applications and products. These include renewable energy technology, electronics and lighting systems, the automotive, metallurgical and nuclear industries (Charalampides et al., 2015; Garcia-Solsona et al., 2014; Guimarães et al., 2016; Ramos et al., 2016). REY are also used in agricultural and animal husbandry products such as fertilisers and feed (Wen et al., 2001; Tommasi et al., 2020). In the last century, the use of anthropogenic REY-chemical complexes (ACC-REY) in medical and pharmaceutical applications has increased significantly. These include the widespread Gd-based contrast agents for magnetic resonance imaging applied in medical diagnostic (see Trapasso et al., 2021 for a comprehensive review on use, fate and effects of Gd as MRI contrast agent), $La_2(CO_3)_3$ used in patients receiving haemodialysis for the reduction of serum phosphorus and La-based compounds which have been proposed for the treatment of patients affected by bone density disorders (Barta et al., 2007; Rogowska et al., 2018).

The global demand for REY is growing rapidly, increasing from 75,500 tonnes (t) in 2000 to 123,100 t in 2016 with an approximated increase of 40% by 2030 (Roskill, 2016, 2019). China is currently, by far, the world's largest REY producer, followed by Australia, USA, Russia, Malaysia and Vietnam (Brown et al., 2013; Golev et al., 2014). As REY are considered 'technology-critical elements', the European Commission defined them as critical raw materials (CRMs) of high strategic importance and identified their supply as one of the greatest European societal challenges (Keersemaker, 2020). To date, several European-level projects have been initiated to assess the potential for REY mining and processing in Europe.

Rising REY production and use has led to an increasing release into the environment (Gwenzi et al., 2018). However, the identification of anthropogenic REY contamination in environmental matrices requires a specific approach owing to their widespread natural occurrence. REY concentrations in environmental samples are commonly presented as normalized to a reference system, in environmental samples typically a shale as an analogue to the REY composition of average upper continental crust. This results in smooth shale-normalized REY ($REY_{SN}$) patterns unless individual REY are anthropogenically enriched (Bau et al., 2018; Censi et al., 2017; Oliveri et al., 2010). REY (micro)contamination is, therefore, shown as positive anomalies (Bau et al., 2018) in their concentration patterns (Bau et al., 1996).

Recent research reported anthropogenically derived REY contamination in various environmental matrices including soil, atmospheric dust, sea and freshwater, with concentrations often orders of magnitude higher than natural geochemical background levels (Bau et al., 2006; Cao et al., 2000). It has also been suggested that ACC-REY chemical-complexes, such as Gd complexes used in diagnostics, can be more stable and bioavailable in environmental matrices compared to geogenic ones (Kulaksiz and Bau, 2013; Liang et al., 2014; Tyler, 2004).

There are several release pathways through which anthropogenically-derived REY can enter the marine environment and/or be transferred across environmental compartments, where they can potentially have adverse effects on organisms and ecosystems. Studies investigating such potential adverse effects have reported a range of impacts, including decreasing survival, reproduction and growth rates as well as alterations in embryonic development and in neural and cardiac activity in freshwater zooplankton, echinoderms and fish (Blaise et al., 2018; Cui et al., 2012; Dubé et al., 2019; Lürling and Tolman, 2010; Zhao et al., 2021). These effects were attributed to inhibition of cellular homeostasis, $Ca2+$ signalling and alteration of gene transcription involved in DNA repair processes. Chronic exposure to REY can also hamper human health, for example by negatively affecting hepatic, respiratory and neural functions (see Gwenzi et al., 2018 for a comprehensive evaluation of the risks exeherted by REY on human health). .

Previous reviews have so far mostly focused on REY detection methods, geochemical processes, application and production, as well as terrestrial distribution, with particular focus on soils and human toxicity (e.g., Adeel et al., 2019; Ascenzi et al., 2020; Ebrahimi and Barbieri, 2019; Fraum et al., 2017; Gwenzi et al., 2018; Telgmann et al., 2013). To our knowledge, no comprehensive multidisciplinary assessment on the sources, distribution, toxicological and ecological effects of REY has yet been performed for the marine environment. As the marine environment is a sink for many contaminants, potentially including anthropogenically realised REY, the current review aims to 1) describe the main geogenic and anthropogenic release pathways of REY to the marine environment; 2) evaluate key factors and processes influencing REY spatial distribution and transfer in the marine environment 3) summarise the available information on REY uptake and accumulation in biota and their eco-toxicological effects; 4) identify and discuss current knowledge gaps and potential future research directions.

## 2. Literature search and selection criteria

### 2.1. Sources and search strategy

The literature search was performed in September 2020 using four different search engines, we examined peer-reviewed papers published up to February 2021: 1) Web of Science (www.webofscience.com); 2) Google Scholar (www.scholar.google.com); 3) PubMed (www.ncbi.nlm.nih.gov/pubmed); 4) ScienceDirect (www.sciencedirect.com).

In a preliminary search, results for the "rare earth elements" and "lanthanoids" in context of this review were compared, with "rare earth elements" providing more comprehensive results. The whole "raw search" comprised five general searches (without restriction of the year of publication, title, abstract and keywords) and four e specific searches. The general searches were performed using the following search strings 1) "rare earth elements AND marine environment"; 2) "rare earth elements" AND "sources AND "marine environment"; 3) "rare earth elements" AND "distribution" AND "marine environment"; 4) "rare earth elements" AND "marine biota". 5) "rare earth elements" AND "effects" AND "marine biota". The more specific searches were performed using the following strings 1) "rare earth elements AND anthropogenic sources AND marine environment" 2) "rare earth elements AND distribution AND marine waters; 3) "rare earth elements AND distribution AND marine sediments" 4) rare earth elements AND toxicological effects* OR ecological effects* AND marine biota. Finally, the reference list of relevant literature reviews on REY were carefully examined to identify any relevant publications missed by the search terms used above.

### 2.2. Literature selection and eligibility criteria

The raw search resulted in more than 10,000 research items and the literature exclusion criteria provided by Moher et al. (2009) were applied. The following items were therefore excluded: not relevant literature according to the review purpose or in another language than English, grey literature, technical reports and conference proceedings. This first selection process resulted in 1341 peer-reviewed research items from 1950 to February 2021. Following a more extensive screening according to the study aims, duplicates and literature lacking of key methodological information and/or data were eliminated, resulting that a total of 125 articles from 1954 to February 2021 (including early on-line publications) were selected as the basis for this review.

### 2.3. Information extraction and qualitative evaluation

Extraction of the information from the final set of research papers was performed by a single reviewer, following categorization of the publication into 1 the following topics: 1) geogenic and anthropogenic sources (n = 40); marine environmental distribution and key responsible mechanisms (n = 49); distribution in marine biota, including different species and tissue (n = 19); ecotoxicological effects on marine biota (n = 17). Where a single publication was applicable to more than one topic, it was categorized only once, but data and results were utilized wherever appropriate.

The key information from each paper was summarised based on the topic, year of publication and geographical distribution (only comprising papers reporting field-based studies) and summarised in Fig. 1 a, b. Of the 120 selected research articles, 69% (n = 90) were published between 2011 and 2021, with 83% (n = 30) of studies the focusing on REY distribution and effects in biota being published during this period. Among the 108 studies performing field evaluations on the selected topics, 32.5% and 35% (n = 29 and n = 32) were performed in Asia and Europe respectively.

## 3. Sources of REY

### 3.1. Natural sources

Geogenic REY are widely distributed in mineral deposits such as Fe–Mn oxides, apatite, bastnaesite, monazite, and other carbonates, phosphates and silicates (Chakhmouradian and Wall, 2012). Generally, HREY reserves are all located within ion-absorption ore deposits, while LREY are mainly contained in carbonatites and alkaline igneous complexes (Khan et al., 2016). Bastnaesite [(Ce, La, Y)$CO_3$F] is a carbonate mineral and serves mostly as source of LREY (particularly for La, Ce) and Y. Monazite and other thorium-bearing phosphate minerals [(Ce, La, Y, Th)$PO_4$] are enriched with REY-oxides and contain predominantly LREY due to low crystallization temperature and pressure. Xenotime ($YPO_4$) is a minor constituent of granitic and gneissic rocks and is enriched in HREY (Khan et al., 2016). Also, co-deposition of REY with natural uranium and thorium has been observed (Melfos and Voudouris, 2012).

Geogenic REY are released and mobilised from mineral deposits as well as from non-mineralised rocks as consequence of natural weathering, erosion and diagenetic and hydrothermal alteration, producing REY fluxes from the Earth's mantle via hydrothermal fluids and from the continental crust via atmospheric dust, volcanic ash, river and porewaters (Consani et al., 2020; Deepulal et al., 2012; Hu et al., 2019; Stichel et al., 2012; Sholkovitz et al., 1999; Xu and Han, 2009). Major geogenic sources of REY and transport pathways to the marine environment are summarised in Fig. 2.

Depending on their origin, REY in aqueous environments are classified in lithogenic and authigenic (Garcia-Solsona et al., 2014). The lithogenic REY fraction comprises particulate matter originating from mechanical erosion of continental/oceanic crust, while the authigenic fraction is formed *in situ* from dissolved REY. Movements of the Earth's crust, volcanic activity, ice melting, hydrothermal activity and groundwater flow can all lead to geogenic REY release, subsequently influence their distribution and concentrations in the marine environment (Caetano et al., 2013; Dia et al., 2000; Janssen and Verweij, 2003; Johannesson et al., 2011; Laukert et al., 2017).

Seasonal atmospheric changes, such as the amount of rainfall, snow precipitation and atmospheric dust fluxes can strongly influence particulate REY deposition and transfer. The atmospheric input of REY and other trace metals to the environment can vary markedly over space and time. Such changes can be rapid for example due to events such as dust storms or explosive volcanic eruptions, or seasonal, and/or multiyear cyclic trends evolving over several millions of years (Bruland and Lohan, 2014; Jickells, 1995; Viehmann et al., 2015; Schier et al., 2021; Zhu et al., 1997).

As it has been studied mainly in terrestrial systems, picks in REY concentrations can be strongly related to seasonal variation in atmospheric dust depositional fluxes and meteorological events. For example Peng et al. (2019), while investigating the annual atmospheric fluxes and seasonal variation of the water-soluble REY fraction in Shihua Cave in North China, found two seasonal deposition peaks, related to intense depositional fluxes of atmospheric dust and low rainfall in summer and autumn. Lower deposition was observed in summer and winter when soil moisture contents change with intense rain- and snowfall.

As for other trace metals, atmospheric inputs could also enhance the deposition of particulate-REY, estimated to be of similar magnitude as riverine inputs (Bruland and Lohan, 2014). Whereas differences in atmospheric fluxes can directly affect the oceanic euphotic zone, fluvial input determine REY enrichment in estuarine and coastal waters

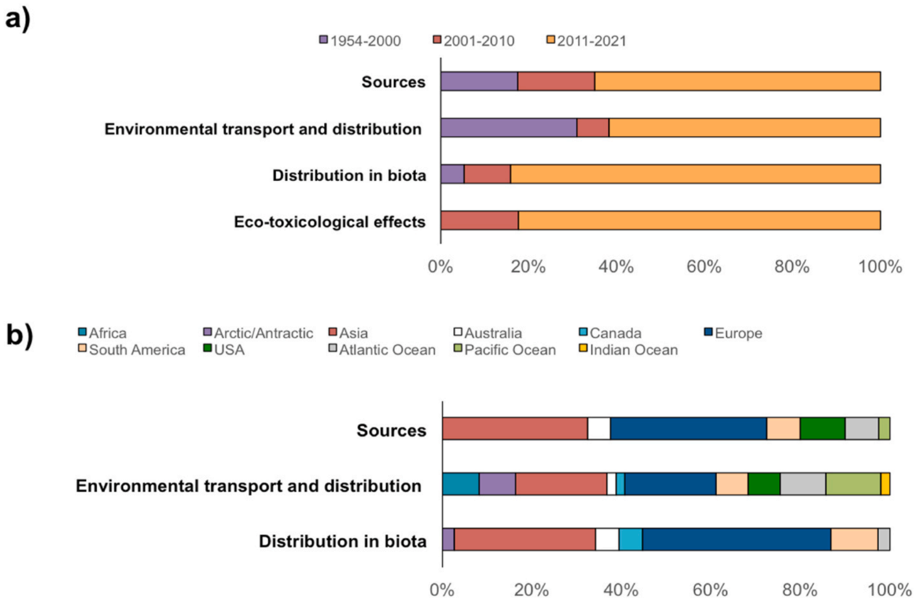

**Fig. 1. a)** Temporal (n = 125 papers from 1954 to February 2021) and **b)** geographical (n = 108, only papers reporting field-based evaluations are considered) distribution of publications on sources and distribution of REY in the marine environment and biota.

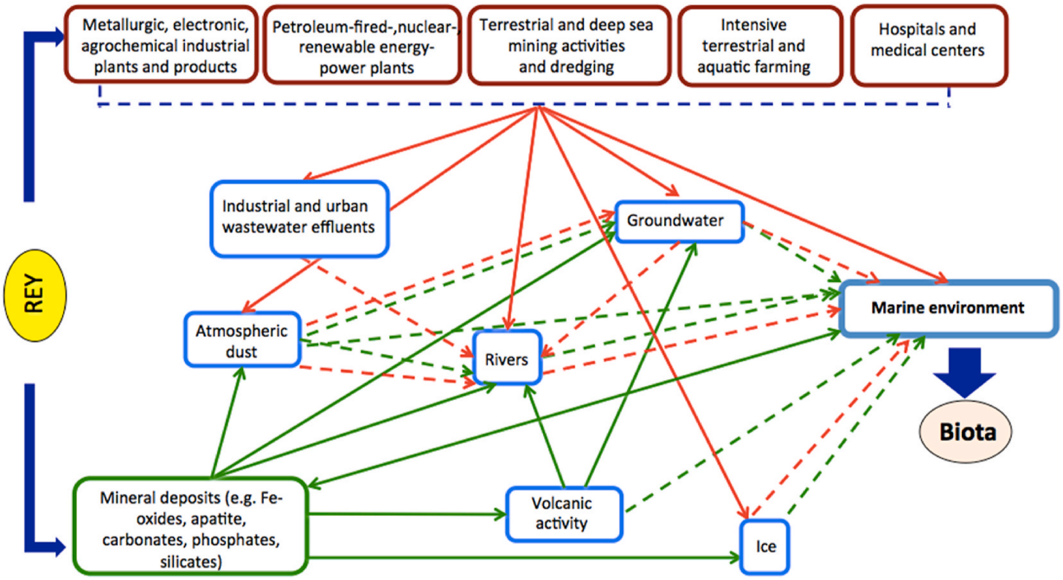

**Fig. 2.** Conceptual representation of the direct (full arrows) and indirect (through a transport-vector; dashed arrows) pathways of transport of REY from geogenic (green) and anthropogenic (red) sources to the marine environment. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

(Jickells, 1995).

### 3.2. Anthropogenic sources

REY are critical raw materials, playing a key role both for traditional industries and, in particular, for technological development of emerging industrial sectors with a focus on enabling technologies. As a result, anthropogenic activities leading to non-geogenic enrichment of (micro) contamination with REY are various and widespread.

The major anthropogenic sources of REY and their transport pathways to the marine environment are summarised in Fig. 2.

REY can be released as particulate matter or dust during processing and use (for a comprehensive review on REY anthropogenic applications refer to Gwenzi et al., 2018). They enter the marine environment via urban and industrial waste-water runoff, rivers and groundwater seepage and as consequence of atmospheric transport and deposition (Brito et al., 2018; Klaver et al., 2014; Morgan et al., 2016; Olmez et al., 1991; Trifuoggi et al., 2018).

In particular, La, Ce, Pr, Nd, Y and Tb can be released as petroleum and coal cracking catalysts and as waste products from different manufacturing process involving the use of mineral precursors such as bastnaesite, monazite and phosphorite. These REY-rich waste products include bottom and fly ashes, airborne particulate matter and atmospheric dust released from oil- and coal-fired power plants and manufacturing, refining, metallurgic and electronic industries and incinerators (Olmez et al., 1991; Fabijańczyk et al., 2019; Funari et al., 2016; Huang et al., 2019; Mao et al., 2014; Pedreira et al., 2004; Suzuki et al., 2010).

REY enrichment in aquatic and atmospheric particulate matter can also occur as effect of the industrial production and use of phosphates fertilisers and animal feeds (Consani et al., 2020; Huang et al., 2019; Otero et al., 2005; Suzuki et al., 2010; Wang et al., 2019; Wen et al., 2001; Zhang and Shan, 2001). Owing to their seemingly growth promoting effects and high abundance in phosphate minerals, certain REY (particularly La, Ce, Pr and Nd) are widely present in chemical fertilisers and animal feeds, where the total REY concentration (ΣREY) may be as high as 1600 ppm (Otero et al., 2005).

Terrestrial and marine mining activities, mine tailing disposal and dredging operations are other significant REY sources, mobilising REY from mineral deposits, soil and sediments (Liang et al., 2014; Mao et al., 2014; Xu et al., 2018).

Of particular environmental concern are the ACC-REY utilized in medical applications. A significant anthropogenic REY source, particularly of Gd, are chemical compounds used as contrast agents in magnetic resonance imaging, which can reach the marine environment through river and waste water outlets (Bau et al., 1996; Bau et al., 1997; Farkas et al., 2020; Hissler et al., 2016; Klaver et al., 2014; Kulaksiz and Bau et al., 2013; Kümmerer and Helmers, 2000; Lawrence et al., 2009; Lerat-Hardy et al., 2019; Song et al., 2017). This can lead to a significant anthropogenic Gd enrichment in coastal waters located within or alongside densely populated areas with highly evolved health care system (Bau et al., 2006) as documented by Kulaksiz and Bau (2007), Nozaki et al. (2000) and Hatje et al. (2016) in the southern North Sea (Germany), Tokyo Bay (Japan) and in San Francisco Bay (USA) respectively.

## 4. Environmental transport and distribution

As other trace metals, the REY exist in the aquatic environment in a variety of physical-chemical forms and can be classified according to the different states in: i) “particulate REY” which are associated to solid particles (>0.2 or 0.45 μm), ii) “dissolved REY” associated to colloids and nanoparticles (<0.2 μm) and iii) " truly dissolved REY” present in the water as single aquo ions or as chemical complexes (Elderfield et al., 1990).

Exchanges between truly dissolved, dissolved and particulate phases occur in oceans and affect spatial distribution, concentrations and bioavailability (Garcia-Solsona et al., 2014). The presence or absence of organic and inorganic nanoparticles and colloids (Merschel et al., 2017), and complexation of REY with organic ligands such as siderophores in freshwaters (Bau et al., 2013) also exert strong control on REY input into seawater.

As typical for particle-reactive elements, individual REY concentrations and the shape of their concentration patterns in the water column and sediments are variable from a both lateral and vertical spatial perspective. This is due to the effects of various factors reflecting different REY sources and processes in the water column, including lithology, geology and climatic conditions in the source region, distance from the coast and exact positio, hydrology of freshwater inputs, presence of anthropogenic inputs and local oceanographic conditions.

**Table 1**
Reported (min-max) water column dissolved REY concentration in open ([a]) and coastal ([b]) (defined as up to 10 km from the coastline and including estuaries and wetlands as suggested in Lavalle et al., 2011) waters. All REY concentrations are reported as ng $L^{-1}$.Values in bold indicate the presence of anthropogenic contamination. b.d.l. = value below the instrumental detection limit. nr = not reported. (Reported values were converted by the authors to uniform the measure unit).

| Area | Region | La | Ce | Pr | Nd | Sm | Eu | Gd | Tb | Dy | Ho | Er | Tm | Yb | Lu | Y | Reference |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **Africa** | South Africa[a] | 1.53–6.39 | 0.16–1.63 | 0.19–1.06 | 0.95–4.7 | 0.18–0.90 | 0.04–0.21 | 0.33–1.24 | 0.04–0.17 | 0.45–1.47 | 0.13–0.37 | 0.48–130 | 0.06–0.18 | 0.39–1.40 | 0.05–0.24 | nr | Garcia-Solsona et al. (2014) |
| **Asia** | Japan[a] | 1.10–5.49 | 0.33–0.77 | 0.16–0.73 | 0.95–3.53 | 0.21–0.69 | 0.06–4.10 | 0.32–1.09 | 0.04–0.17 | 0.40–1.37 | 0.09–0.37 | 0.34–1.35 | 0.05–0.20 | 0.29–1.47 | 0.03–0.24 | 6.35–20 | Alibo and Nozaki (1999) |
|  | Japan[b] | 0.87–30.1 | 0.43–24.1 | 0.33–4.04 | 1.86–12.2 | 0.56–3.35 | 0.12–1.90 | **1.32–24.4** | 0.11–2.18 | 1.07–0.25 | 0.36–9.04 | 1.49–40.3 | 0.28–7.42 | 2.30–64 | 0.33–14.7 | 17.4–192 | Nozaki et al. (2000) |
| **Europe** | Greece[a] | 2.08–0.25 | 1.50–81.1 | 0.37–4.47 | 1.75–19.5 | 0.46–4.11 | 0.13–1.08 | 0.72–5.41 | 0.14–0.92 | 1.07–4.63 | 0.29–1.09 | 1.05–3.33 | 0.15–0.27 | 1.09–1.44 | 0.19–0.40 | 16.4–39.6 | Bau et al. (1997) |
|  | Germany[b] | 0.82–9.18 | 1.18–14.5 | 0.21–1.79 | 0.96–7.07 | 0.27–1.83 | 0.07–0.50 | **0.03–9.44** | 0.06–0.46 | 0.19–3.38 | 0.11–0.75 | 0.1–2.43 | 0.01–0.37 | 0.1–2.39 | 0.01–0.42 | nr | Paffrath et al. (2020) |
|  | Spain[b] | 6.27–72.9 | 6.62–139 | 0.63–18.3 | 2.64–78.3 | 0.67–12.6 | b.d.l-2.43 | 0.46–15.4 | b.d.l.-2.86 | 0.46–15.4 | b.d.l-2.86 | 0.61–6.15 | 0.12–1.18 | 0.67–3.85 | b.d.l.-0.25 | 15.3–109 | Cánovas et al. (2020) |
| **South America** | Brasil[b] | 2.64–9.31 | 3.50–30.1 | 0.60–5.64 | 2.88–21.8 | 0.63–4.81 | 0.27–1.17 | **1.12–5.98** | 0.15–3.81 | 1.32–3.90 | 0.34–0.89 | 1.07–2.64 | 0.15–0.38 | 1–2.58 | 0.17–0.47 | nr | Andrade et al. (2020) |
| **USA** | Alaska[b] | 2.04–18.9 | 0.40–32.2 | 0.20–4.28 | 0.97–18 | 0.12–1.02 | 0.05–1.03 | 0.30–4.70 | 0.03–0.73 | 0.33–4.50 | 0.10–1 | 0.36–2.89 | 0.04–0.42 | 0.26–2.58 | 0.03–0.42 | nr | Haley et al. (2014) |
|  | Oregon[a] | 2.36–9.05 | 0.25–6.39 | 0.34–1.44 | 1.53–6.24 | 0.28–1.41 | 0.08–0.46 | 0.04–1.69 | 0.09–0.30 | 0.60–2.11 | 0.15–0.60 | 0.58–1.99 | 0.08–0.35 | 0.48–2.29 | 0.07–0.43 | nr | Abbott et al. (2015) |
| **Atlantic Ocean** | South Atlantic[a] | 2.04–18.9 | 0.40–32.2 | 0.20–4.28 | 0.97–18 | 0.12–1.02 | 0.05–1.03 | 0.30–4.70 | 0.03–0.73 | 0.33–4.50 | 0.10–1 | 0.36–2.89 | 0.36–2.89 | 0.26–2.58 | 0.03–0.42 | nr | Zheng et al. (2016) |
|  | South Atlantic[a] | 2.12–3.61 | 0.71–2.19 | nr | 2.23–3.09 | 0.49–0.81 | 0.12–14.4 | 0.79–1.14 | nr | 0.83–0.97 | nr | 0.74–0.85 | nr | 0.68–0.82 | 0.08–0.12 | nr | Sholkovitz et al., 1994 |
| **Pacific Ocean** | South Pacific[a] | 2.85–3.19 | 3.50–3.98 | 0.55–0.81 | 2.71–3.72 | 0.54–0.72 | 0.16–0.22 | 0.58–0.79 | 0.07–0.12 | 0.55–0.78 | 0.11–0.18 | 0.35–0.48 | 0.03–0.06 | 0.24–0.46 | 0.03–0.07 | 8.07–9.66 | Molina-Kescher et al. (2018) |
|  | North Pacific[a] | 0.61–2.60 | 0.56–4.74 | 0.11–1.55 | 0.56–6.79 | 0.09–0.63 | 0.03–0.41 | 0.20–2.44 | 0.03–0.17 | 0.27–2.88 | 0.06–0.69 | 0.28–1.57 | 0.03–0.28 | 0.18–2.01 | 0.01–0.38 | 5.01–32.3 | Tazoe et al., 2011 |
|  | Westernpacific[a] | 1.15–6.97 | 0.78–2.56 | 0.22–1.41 | 1.04–5.21 | 0.16–1.32 | 0.03–0.31 | 0.23–1.51 | 0.03–0.20 | 0.26–1.69 | 0.04–1.69 | 0.16–1.19 | 0.01–0.17 | 0.10–1.21 | 0.01–0.21 | nr | Deng et al. (2017) |
| **Indian Ocean** | SW Indian Ocean[a] | nr | nr | nr | 0.98–4.41 | nr | nr | nr | nr | nr | nr | nr | nr | nr | nr | nr | Amakawa et al. (2019) |
| **Arctic** | Laptev sea[a, b] | 1.75–72.7 | 0.62–82.6 | 0.38–19 | 1.86–82.3 | 0.41–17.6 | 0.10–3.43 | 0.62–17.5 | 0.1–2.49 | 0.81–15.1 | 0.21–3.28 | 0.73–9.36 | 0.09–1.33 | 0.63–8.83 | 0.10–1.53 | 10.8–103 | Laukert et al. (2017) |

Data on REY concentrations in seawater and sediments are extremely variable from a spatial perspective, making the establishment of global trends difficult. Reported concentrations of REY in seawater and sediments range from a factor of $10^{-2}$ to $10^{2}$ and $10^{-2}$ to 10 in seawater and sediments respectively, with concentrations decreasing from LREY to HREY, and generally higher concentrations in coastal areas compared to open oceans (Tables 1 and 2).

The concentration, distribution and persistence of REY in the water column and their vertical transport is largely dominated by complexation with functional groups at the surface of suspended particles, colloids and nanoparticles and by the fractionation between the truly dissolved and dissolved/particulate REY (Bertram and Elderfield, 1993; Elderfield et al., 1988; Sholkovitz et al., 1994 Watkins et al., 1995). REY fractionation is, in turn, influenced by a variety of environmental factors including pH, salinity, temperature, redox level, and availability of ligands, particularly of phosphate and carbonate, which can promote changes in REY state and determine their permanence in a specific compartment (Andrade et al., 2020; Byrne et al., 1988; Byrne and Kim, 1993; Byrne and Sholkovitz, 1996; Goldstein and Jacobsen, 1988; Xu and Han, 2009). Particularly, the vertical transport of REY is affected by scavenging, defined as the combined processes of surface adsorption followed by particle settling (Goldberg, 1954; Turekian, 1977), promoting REY sequestration to sediments (Andrade et al., 2020; Byrne and Kim, 1990; Byrne and Sholkovitz, 1996; Casse et al., 2019).

Generally, dissolved REY concentrations are increasing with depth, accompanied by enrichment of HREY compared to LREY, which conversely, tend to be sequestrated in sediments at higher rates (Abbott et al., 2015; Alibo and Nozaki, 1999; Garcia-Solsona et al., 2014; Haley et al., 2014). This is related to a higher lithogenic supply of LREY than HREY, surface removal due to scavenging and deep-remineralisation accompanied by high sedimentation rates. This trend may be inverted in coastal areas, where continuous anthropogenic REY inputs can cause enrichment in surface waters. Variations in REY concentration can also be associated to tidal fluctuations porewater flux and/or freshwater seepage (Andrade et al., 2020; Cánovas et al., 2020; Tazoe et al., 2011), while in open oceans, REY concentrations are strongly affected by oceanic circulation, water mass mixing and biogeochemical cycling (Amakawa et al., 2019; Alibo and Nozaki, 1999; Ye et al., 2019; Zheng et al., 2016).

Ice-related processes have been also identified as factor influencing dissolved REY concentrations in the water column. Particularly ice-melting seems to be a mechanism leading to REY dilution in surface waters of Arctic estuaries, resulting in increasing REY concentrations from the surface to the bottom (Laukert et al., 2017). This is further amplified by scavenging of REY by rock flour in organics-poor arctic rivers and estuaries (Tepe and Bau, 2015, 2016). In coastal areas, these trends may also be inverted due to tidal fluctuations, porewater flux and/or fresh water inputs of organic matter and particularly due to continuous anthropogenic REY inputs which can cause REY enrichment both in water and sediments (Andrade et al., 2020; Casse et al., 2019; Cánovas et al., 2020; Consani et al., 2020; Fiket et al., 2017; Freslon et al., 2014; Hu et al., 2019; Tazoe et al., 2011).

After settling to sediments, REY can, depending on local microbiological activity, sedimentation rates, and hydrodynamic conditions, either undergo mineralisation processes and thereby being sequestrated, or can be mobilised and resuspended (Bruland and Lohan, 2014; Censi et al., 2014; Pattan et al., 1995). Several studies report that REY sediment patterns are highly related to the sediment properties such as grain-size (granulometry). The presence of high REY concentrations, particularly LREY, has been associated with the occurrence of fine-grained particulates such as sand and silt fractions compared to coarse-grained sands and gravel (Astakhov et al., 2019; Brito et al., 2018; Caetano et al., 2009; Tranchida et al., 211; Trifuoggi et al., 2018). It has been suggested that this can reflect the higher adsorption capacity of smaller particles featuring a higher surface to volume ratio (Sholkovitz et al., 1999).

**Table 2**
Reported REY concentration (min-max) in marine sediments from open ([a]) and coastal ([b]) (defined as up to 10 km from the coastline and including estuaries and wetlands as suggested in Lavalle et al., 2011) areas. All REY concentrations are reported as mg kg$^{-1}$ (measured on dry weight of sediment). Values in bold indicate the presence of anthropogenic contamination. nr = "not reported" in the source paper. (Reported values were converted by the authors to uniform the measure unit).

| Area | Region | La | Ce | Pr | Nd | Sm | Eu | Gd | Tb | Dy | Ho | Er | Tm | Yb | Lu | Y | Reference |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **Africa** | Egypt[b] | 0.6–20.1 | 1.4–25.2 | 0.2–3.2 | 0.7–35.3 | 0.1–1.7 | 0.1–1 | 0.1–9 | 0.1–.8 | 0.1–0.9 | 0.1–1.2 | 0.1–4.1 | 0.1–0.7 | 0.1–4.3 | 0.1–0.7 | nr | El-Taher et al. (2019) |
|  | Namibia; South Africa[b] | 3–93 | 6–352 | 0.8–23.5 | 3–98 | 0.6–6.4 | 0.2–5 | 0.7–26.2 | 0.09–3.6 | 0.6–22 | 0.1–4.4 | 0.4–13 | 0.08–0.4 | 0.4–11.4 | 0.08–0.4 | nr | Orani et al. (2018) |
|  | Namibia; South Africa[b] | 0.6–234 | 1.4–565 | 0.1–57.6 | 0.7–238 | 0.1–50.8 | 0.04–11.4 | 0.1–55.3 | 0.02–8.3 | 0.1–50.6 | nr | 0.07–29.4 | nr | 0.05–26.8 | nr | nr | Watkins et al. (1995) |
| **Asia** | Siberia[a,b] | 13–45 | 26 –88 | 3.1–9.4 | 12 –34 | 2.4–7 | 0.5–1.3 | 1.9–5.4 | 0.2–0.8 | 1.5 –4.4 | 0.2–0.8 | 1.0–2.6 | 0.1–0.3 | 0.6–2.6 | 0.09–0.3 | 7–23 | Astakhov et al. (2019) |
|  | China[a] | 31.8–37.3 | 61.6–70.8 | 6.8–8.3 | 26.2–29.8 | 4.8–5.7 | 1–1.2 | 4.3–5.3 | 0.6–0.8 | 3.8–4.5 | 0.7–0.8 | 2.2–2.6 | 0.3–0.3 | 2.1–2.4 | 0.3–0.3 | nr | Chen et al. (2020) |
|  | China[b] | 42.3–66.7 | 82.5–132 | 9.08–14.2 | 32–52 | 5.9–9.5 | 0.9–1.6 | 5.3–8.6 | 0.7–1.3 | 3.98–6.54 | 0.789–1.32 | 2.5–3.8 | 0.3–0.6 | 2.4–4.0 | 0.3–0.6 | nr | Hu et al. (2019) |
|  | Saudi Arabia[b] | 13.9–52.3 | 27.7–102.8 | 2.77–10 | 11–38.4 | 1.9–6.9 | 0.5–1.6 | 1.3–5.6 | 0.1–0.8 | 1–4.9 | 0.2–1 | 0.5–2.8 | 0.08–0.3 | 0.5–2.3 | 0.08–0.3 | nr | Deepulal, et al. (2012) |
|  | Saudi Arabia[b] | 2.9–9.7 | 3.1–10.4 | nr | nr | 1.7–3.7 | 2.2–4.6 | nr | 6.1–9.9 | nr | nr | **nr** | nr | 2.1–4.5 | 0.9–1.2 | nr | El-Taher et al. (2018) |
|  | Korea[a] | 6-163 | 16-80 | 4.8–50 | nr | 3.9–47 | 1.5–12.4 | 5.9–53 | nr | 6-50 | 1.2–10.8 | 3.5–29.1 | 0.49–4 | 3.3–26.1 | 0.49–4 | nr | Jung et al. (1998) |
|  | Malaysia Singapore[b] | 13.9–27 | 10.4–43 | nr | 15.3–39.9 | 3.5–10.1 | 0.6–5 | nr | nr | 0.5–3.7 | nr | nr | nr | 1.3–2.5 | nr | nr | Khadijeh et al. (2009) |
|  | Malaysia Singapore[b] | 7.3–73.5 | 24.5–77 | nr | nr | 1.7–12.7 | 0.2–0.8 | nr | 0.1–0.8 | 3.02–4.08 | nr | nr | nr | 1.5–3.6 | nr | nr | Yusof et al. (2001) |
| **Australia** | Western Australia[b] | 1-321 | 1.9–971 | 0.2–67 | 0.7–269 | 0.1–46 | 0.1–10 | 0.1-54 | 0.1–7.4 | 0.3–34 | 0.1–6.2 | 0.2–16 | 0.1–2.2 | 0.1–13 | 0.1–1.9 | 0.3–170 | Xu et al. (2018) |
| **Canada** | Eastern Canada[b] | 20.9–37.2 | 41.6–73.7 | 4.38–9 | 16.08–34.2 | 2.8–5.1 | 0.6–1.6 | 2.5–5.4 | 0.3–0.7 | 1.7–4.3 | 0.3–0.8 | 1.09–2.5 | 0.1–0.3 | 1.04–2.3 | 0.1–0.3 | nr | Casse et al. (2019) |
| **Europe** | Portugal[b] | **3.7–74** | **7.5–87** | **0.9–11** | **3.6–43** | **0.7–7.3** | **0.1–1.4** | 0.5–5.8 | 0.07–0.8 | 0.4–4.5 | 0.08–0.8 | 0.23–2.3 | 0.03–0.3 | 0.2–2.4 | 0.03–0.3 | 2.4–36 | Brito et al. (2018) |
|  | Spain[b] | 12-39 | 21-70 | 2.9–9.1 | 10-30 | 2–6 | 0.5–1.2 | 2–5.7 | 0.2–0.5 | 1.4–3.8 | 0.2–0.5 | 0.7–2 | 0.08–0.2 | 1–1.9 | 0.07–0.21 | nr | Caetano et al. (2009) |
|  | Italy[a] | 4.6–13.6 | 11.01–29.9 | 1.1–3.3 | 4.3–13.3 | 1.1–2.8 | 0.1–0.6 | 1.2–2.8 | 0.1–0.4 | 0.6–2.1 | 0.1–0.4 | 0.3–1.08 | 0.04–0.3 | 0.2–0.8 | 0.03–0.1 | 2.5–10.9 | Censi et al. (2014) |
|  | Italy[b] | **1.9–6.8** | **5.1–16.4** | 0.7–2.6 | 4.02–11.5 | 1–3.1 | 0.3–0.6 | **1.2–2.7** | 0.2–0.4 | 1.08–2.2 | 0.2–0.5 | 0.5–0.9 | 0.06–0.1 | 0.3–0.7 | 0.05–0.1 | **5.3–14.4** | Consani et al. (2020) |
|  | Italy[a,b] | 15.2–34.4 | 33.1–68.7 | 3.9–8.5 | 15.5–32.7 | **2.9–6.3** | **0.6–1.4** | **2.8–6.2** | nr | 2.1–3.6 | nr | 0.8–1.6 | nr | 0.7–1.1 | 0.1–0.1 | 8.7–12.8 | Tranchida et al. (2011) |
|  | Italy[b] | **0.3–48.2** | **12.7–98.5** | 0.2–4.8 | 0.7–30.9 | **0.1–2.6** | **0.07–0.4** | 0.2–2.5 | 0.04–0.4 | 0.1–1.9 | 0.03–0.2 | 0.08–1 | 0.02–0.1 | 0.09–0.6 | 0.02–0.1 | **0.3–14.8** | Trifuoggi et al. (2018) |
|  | Croatia[b] | 6.4–38.2 | 10.3–80.3 | 1.42–7.94 | 6.7–35.2 | 1.2–5.8 | 0.2–1.2 | 1.05–4.8 | 0.1–0.7 | 0.9–3.7 | 0.1–0.7 | 0.4–2.3 | 0.06–0.2 | 0.4–2.16 | 0.05–0.2 | nr | Fiket et al. (2017) |
| **USA** | Texas[b] | 25.2–48.9 | 47.5–107 | 7.35–13.3 | nr | 5.1–11.9 | nr | nr | nr | 3.02–5.2 | 0.6–1.4 | 1.4–2.9 | 0.2–0.5 | 1.4–3.9 | nr | nr | Ravichandran (1996) |
| **Indian Ocean** | Wharton Basin[a] | 3–177.6 | 0.6–180.7 | nr | 2.6–186 | 0.7–37.3 | 0.2–10.9 | 0.7–44 | nr | 0.6–22 | 0.1–10 | 0.4–29.3 | nr | 0.5–23.1 | 0.1–1.9 | 12–386 | Pattan et al. (1995) |
| **Pacific Ocean** | Takuyo seamount NW Pacific[a] | 69.8–354 | 10.9–219 | 41.4–981 | 9.05–229 | 2.06–55 | 8.4–249 | 1.3–37.9 | 7.7–235 | 1.6–48.7 | 4.5–136 | 0.6–18.4 | 4.2–114 | 0.6–16.8 | 39.5–1525 | 43.2–739 | Tanaka et al. (2020) |
| **Atlantic Ocean** | Mid-Atlantic ridge[a] | 4.09–12.5 | 7.3–24.1 | 1.3–5.4 | 6.6–25.1 | 1.3–6.5 | 0.3–1.8 | 1.4–6.2 | 0.2–0.9 | 1.2–6.1 | 0.2–1.1 | 0.6–3.1 | 0.09–0.2 | 0.5–2.9 | 0.08–0.4 | 7.5–22.9 | Li et al. (2015) |
| **Arctic Ocean** | Alfa ridge[a] | 24-40 | 50–105 | 6–10 | 22-36 | 4–7 | 1–2 | 4–7 | 0.5–1 | 3–6 | 0.6–1.1 | 2–3 | 0.2–0.4 | 2–3 | 0.2–0.4 | nr | Ye et al. (2019) |

**Table 3**

∑REY concentrations in marine organisms collected in wild or semi-wild conditions. Values are, depending on the reported source format, as either mean ± SD of ΣREY ($^{\text{ł}}$), mean (min-max) of ΣREY ($^{\text{Ŧ}}$), the sum of mean individual REY ($^{\phi}$). All concentrations are reported as μg $kg^{-1}$ ($^{a}$dry weight or $^{b}$wet weight). nr = value/information not reported or not clearly specified in the source paper.

| Country | Cohort | Species/Order | Tissue $^{a/b}$ | ∑REY | REY occurrence | Reference |
|---|---|---|---|---|---|---|
| **Australia** | Corals | *Porites* spp. | $^{a}$nr | $^{\text{ł}}$174.78 ± 47.71 | Y > Ce > La > Nd > Gd > Yb > Dy > Er > Pr > Sm > Ho > Eu > Lu > Tb > Tm | Leonard et al. (2019) |
| **China** | Molluscs | *Eustrombus gigas; Trochus niloticus; Harpago chiragra; Tridacna Squamosa* | $^{a}$nr | $^{\text{Ŧ}}$351.202 (30.407–1481.776) | La > Ce > Nd > Pr > Sm > Gd > Dy > Er > Yb > Eu > Ho > Tb > Tm > Lu | Li et al. (2015) |
| | | *Moerella iridiescens; Ruditapes philippinarum* | $^{a}$nr | $^{\text{ł}}$442.94 ± 53.23 | Ce > La > Nd > Y > Pr > Gd > Sm > Dy > Er > Yb > Eu > Ho > Tb > Tm > Lu | Wang et al., (2019) |
| | Crustaceans | *Dromia dehaani; Paguridae; Podophthalmus virgil; Carpilus convexu* | $^{a}$nr | $^{\text{Ŧ}}$372.58 (103.06–864.16) | La > Ce > Nd > Gd > Dy > Sm > Pr > Er > Yb > Ho > Eu > Tb > Tm > Lu | Li et al. (2015) |
| | | *Scylla serrata;*<br>*Penceus penicillatus* | $^{a}$nr | $^{\text{ł}}$189.48 ± 153.13 | La > Ce > Nd > Gd > Dy > Pr > Sm > Er > Yb > Y > Eu > Ho > Tb > Tm > Lu; Ce > La > Nd > Y > Pr > Gd > Sm > Dy > Er > Yb > Eu > Ho > Tb > Tm > Lu | Wang et al. (2019) |
| | Fish | Over 30 different species | $^{a}$nr | $^{\text{Ŧ}}$13.345 (4.114–44.721) | Ce > La > Nd > Gd > Pr > Sm > Eu > Dy > Yb > Er > Tb > Ho > Tm > Lu | Li et al. (2015) |
| | | | $^{b}$muscle | $^{\text{ł}}$21.01 ± 8.17 | LREY > HREY | Yang et al. (2016) |
| | | | $^{a}$nr | $^{\text{ł}}$10.27 ± 3.60 | Ce > Nd/La/>Nd/La > Gd > Tb > Y > Dy > Pr > Sm > Er > Yb > Eu; Ho > Tm > Lu | Wang et al. (2019) |
| **Japan** | Macroalgae | *Ecklonia cava;*<br>*Delisea fimbriata;*<br>*Ptilonia okadai;*<br>*Ulva fasciata;*<br>*Codium fragile* | $^{a}$blade $^{a}$stipe<br>$^{a}$nr<br>$^{a}$nr<br>$^{a}$holdfast<br>$^{a}$nr | $^{\phi}$121; 50<br>$^{\phi}$175;<br>$^{\phi}$215;<br>$^{\phi}$548;<br>$^{\phi}$235 | Ce > La > Nd > Gd > Dy > Yb > Pr > Sm; >Er > Ho > Eu<Lu>Tb > Tm;<br>Ce > Nd > La > Gd > Sm; Dy > Pr > Er > Yb > Eu > Ho > Tb > Tm > Lu;<br>Ce > Nd > La > Gd > Dy > Sm > Pr > Er > Yb > Eu > Ho > Tb > Tm > Lu<br>Ce > La > Nd > Gd > Dy > Sm > Pr > Er > Yb > Eu > Ho > Tb > Lu > Tm;<br>Ce<Nd>La > Eu > Dy; Gd > Sm > Pr > Yb > Ho > Er > Tb > Lu > Tm; | Fu et al. (2000) |
| | Molluscs | *Ruditapes philippinarum;*<br>*Mercenaria stimpsoni;*<br>*Mytilus galloprovincialis;*<br>*Mactra veneriformis;*<br>*Phacosoma japonicum* | $^{a}$soft tissue | $^{\text{ł}}$2451.95 ± 876.66 | Ce > Nd > La > Pr/Sm/Gd/Dy > Er/Eu/Yb > Ho/Tb > Tm/Lu | Akagi & Edanami (2017) |
| | | | $^{a}$shells | $^{\text{ł}}$301484.4 ± 202179.5 | Ce > La > Nd > Pr/Gd > Pr/Gd > Sm/Dy > Sm/Dy > Er > Yb > Eu/Ho > Eu/Ho > Tb > Tm/Lu > Tm/Lu | |
| **Malaysia** | Macroalgae | *Padina* sp. | $^{a}$apical and middle part tissue | $^{\text{ł}}$1846.45 ± 14990.14 | LREY > HREY | Mashitah et al. (2012) |
| **Canada** | Blue mussel | *Mytilus edulis* | $^{a}$whole soft tissue | $^{\text{ł}}$5170 ± 810 | Ce > La > Nd > Y > Pr > Sm > Gd > Dy > Er > Eu > Yb > Tb > Ho > Tm, Lu | MacMillan et al. (2017) |
| | Sea urchin | Echinoderm | $^{a}$gonads | $^{\text{ł}}$2210 ± 990 | La > Ce > Y > Nd > Pr > Gd > Sm > Dy > Er > Yb > Eu > Tb > Ho > Tm > Lu | |
| | Common eider | *Somateria mollissima* | $^{a}$muscle | $^{\text{ł}}$8±693 | Ce > La > Y,Nd > Sm,Gd > Pr > Eu,Dy,Yb | |
| | | | $^{a}$liver | $^{\text{ł}}$46 ± 43 | La > Ce > Nd > Pr > Y > Sm > Gd > Eu, Dy,Yb | |
| | Ringed seal | *Phoca hispida* | $^{a}$muscle | $^{\text{ł}}$3±19 | Ce > La,Nd,Y > Sm,Gd | |
| | | | $^{a}$liver | $^{\text{ł}}$115 ± 180 | Ce > La > Nd > Pr > Y > Sm > Gd > Eu > Dy > Tb; Er; Yb | |
| **Chile** | Pilot whale | *Globicephala melas* | $^{a}$blubber | $^{\text{ł}}$5510 ± 1920 | Only Ce analysed | Garcia-Cegarra et al. (2020) |
| **Cuba** | Indopacific lionfish | *Pterois* spp. | $^{b}$muscle | $^{\text{ł}}$16 ± 2.2; | HREY > LREY | Squadrone et al. (2020) |
| | | | $^{b}$liver | $^{\text{ł}}$18 ± 1.1 | HREY > LREY | |
| | | | $^{b}$kidneys | $^{\text{ł}}$53 ± 6 | LREY > HREY | |

(*continued on next page*)

**Table 3** (*continued*)

| Country | Cohort | Species/Order | Tissue $^{a/b}$ | ∑REY | REY occurrence | Reference |
|---|---|---|---|---|---|---|
| **France** | Zooplankton | | $^{a}$pool of individuals | $^{ł}$17.72 ± 3.78 | LREY > HREY | Strady et al. (2015) |
| **Italy** | Macroalgae | Clorophyta, Ochorophyta and Rhodophyta: *Codium bursa; Flabellia petiolata;Caulerpa racemosa* and *cylindracea; Padina pavonica; Halimeda tuna; Halopteris filicina; scoparia; Ganonema farinosum; Dyctyota dichotoma; Peyssonnelia squamaria; Laurencia obtusa; Cystoseira* spp.; *Dudresnaya verticillata; Acetabularia acetaulum; Phyllophora crispa* | $^{a}$nr | $^{ł}$7900 ± 4600 –$^{ł}$22000 ± 4900 | LREY > HREY | Squadrone et al. (2017) |
| | | | | $^{ł}$11300 ± 6300 | LREY > HREY | Squadrone et al. (2018) |
| | | | $^{a}$nr | $^{ł}$12000 ± 6900 | LREY > HREY | Squadrone et al. (2019a) |
| | Zooplankton | | $^{a}$nr | $^{ł}$120 ± 20 | LREY > HREY | |
| | Molluscs | Mussels; clams; oysters | $^{a}$whole soft tissue | $^{ł}$160 ± 79 | LREY > HREY | |
| | Fish | Mullet; redfish; mackerel; hake; | $^{a}$muscle | $^{ł}$210 ± 23 | LREY > HREY | |
| | | *Mallotus villosus* | $^{nr}$nr | $^{ł}$5.7 ± 0.3–150 ± 21 | LREY > HREY | Squadrone et al. (2019b) |
| | Penguins | *Sphenicus humboldti* | feathers | $^{ł}$160 ± 23–160 ± 29 | LREY > HREY | |
| **Mid-Atlantic Ridge; Ireland; Germany** | Mussels | *Bathymodiolus spp; Mytilus edulis* | shells | 355.03 ± 209.13 | LREY > HREY | Bau et al., (2010); |
| **Germany** | Mussels | *Mytilus edulis* | shells | $^{ł}$55203.33 ± 7487.61 | Y > La > Ce > Nd > Gd > Pr > Sm > Dy > Er > Eu > Yb > Tb > Ho > Lu | Ponnurangam et al. (2016) |
| **Spain** | Mussels | *Mytilus galloprovincialis* | $^{a}$soft tissue | $^{ł}$556.92 ± 182.68 | Ce > La > Nd > Y > Pr > Gd > Sm > Dy > Er > Yb > Eu > Ho > Lu > Tm | Costas-Rodríguez et al. (2010) |
| **Southern Baltic Sea** | Fish | *Cluepea harengus membras;* | $^{b}$muscle | $^{\phi}$57 | Pr > La > Ce > Sm > Nd > Tb > Y > Dy > Tm ≫ Yb > Er > Gd > Eu > Ho | Reindl et al. (2021) |
| | Grey Seal | *Halichoerus grypus grypus;* | $^{a}$fur | $^{\phi}$489; | Ce > Nd > Y > La | |
| | | | $^{a}$faeces | $^{\phi}$676 | Ce > La > Nd > Y > Gd > Sm> | |
| **Antarctic Peninsula** | Fish | *Notothenia rosii* | $^{b}$muscle | $^{\phi}$540 | Y > Dy > Nd > La ≫ Eu > Ce > Pr > Gd > Tb > Yb | |
| | Southern elephant seal | *Mirounga leonine* | $^{a}$fur | $^{\phi}$10010 | Ce > Nd > Y > La > Gd > Pr > Sm > Dy > Yb > Er > Eu > Ho > Tb > Tm > Lu | |
| | | | $^{a}$faeces | $^{\phi}$83600 | Ce > Nd > Y > La > Gd > Pr > Sm > Dy > Er > Yb > Eu > Tb > Ho > Tm > Lu | |

## 5. REY interaction with biota and biological effects

### 5.1. Occurrence in biota and tissue distribution

The occurrence and accumulation of total (∑REY) or individual REY have been, so far, less studied compared to the abiotic compartments and REY have mainly been quantified in macroalgae, benthic invertebrates including bivalves, echinoderms, crustaceans and fish (Akagi & Edanami, 2017; Bau et al., 2010; Fu et al., 2000; Li et al., 2015; MacMillan et al., 2017; Mashitah et al., 2012; Reindl et al., 2021; Ponnurangam et al., 2016; Squadrone et al., 2017, 2018, 2019a,b, 2020; Wang et al., 2019; Yang et al., 2016) (Table 3).

Even if a direct comparison among studies is not always possible due to the use of different units and data reporting (as Σ or individual REY; in tissue wet or dry weight), studies commonly report that LREY occur at higher concentrations in organisms compared to HREY with La, Ce and Nd having the highest concentration (Table 3). Organisms at lower trophic levels such as macroalgae and invertebrates, seem to generally exhibit higher REY concentrations (up to 4 orders of magnitude) compared to species at higher trophic positions (e.g., fish, mammals and birds). This suggests that organisms feeding directly from the water and sediments may be more prone to take up REY and that there is limited potential for REY to biomagnify, but that they rather biodilute in food chains, a pattern that is observed for several other elements (for example Sun et al., 2020; Ciesielski et al., 2016). Different level of REY could be related to different uptake rates and exposure according to the feeding strategy and/or specific habits (e.g. mobility, localisation in different compartments) of different organisms. In fact, sessile benthic organisms, such as macroalgae and non-selective filter-feeders (for example mussels, clams and oysters), which can be continuously and directly exposed to REY from both the water column and sediments, exhibit the highest level of REY (up to 22 mg kg$^{-1}$ of dry weight in macroalgae; Table 3) and similar patterns as the seawater they grew and feed in (Bau et al., 2010; Ponnurangam et al., 2016; Squadrone et al., 2017).

In addition, organisms at higher trophic positions which have more effective metabolic mechanisms to regulate organismal concentrations of metals (Liu et al., 2019), could egest REY at higher rates compared to species at lower positions. For example, Reindl et al. (2021) reported relatively high levels of REY in seals' faeces, about 1–2 order of magnitude higher compared to the edible tissue of their prey, thereby also confirming that limited levels of biomagnification of REY occur in natural populations. REY have been also previously described to strongly partition from seawater into calcite, substituting for Ca$^{2+}$, with their partition coefficients decreasing with atomic number from LREY to HREY (Zhong and Mucci, 1995). Therefore, this Ca$^{2+}$ substitution mechanism could also play a role in higher REY accumulation in lower trophic levels in marine species, as many lower trophic marine organisms actively incorporate Ca$^{2+}$ during mineralisation processes to form

**Table 4**
Exposure studies on marine organisms reporting biological effects after exposure to individual or mixtures of REY. Values are, depending on the reported source format, as either µg $L^{-1}$ ($^{\dagger}$) or µM ($^{\phi}$).

| Cohort | Species | Life stage | Tested REY compound | Effect concentration (EC) | Effects | Reference |
|---|---|---|---|---|---|---|
| **Algae** | *Skeletonema pseudocostatum* | | $(La–Y)Cl_3$ or $(NO_3)_3$ | $^{\phi}EC_{50}$ La–Lu = 28.53–30.34; Y = 43.21;<br>$^{\phi}EC_{50}$ = 30.32 | Inhibition of growth | Tai et al. (2010) |
| **Crustaceans** | *Artemia salina* | adult | $La^{3+}$; $Nd^{3+}$; $Sm^{3+}$ | $^{\dagger}EC_{50} > 10^5$<br>$^{\dagger}10^4 < EC_{50} > 10^5$ | Moderate toxicity (La, Nd, La + Nd, La + Sm, Nd + Sm); low toxicity (Sm) | Bergsten-Torralba et al. (2020) |
| **Molluscs** | *Mytilus galloprovincialis* | adult | $Nd^{3+}$ | $^{\dagger}$5-40; | Stimulation of metabolic and antioxidant enzyme activity; decrease in energy reserves | Freitas et al. (2020a) |
| | | | | $^{\dagger}$2.5-10 | Stimulation of biotransformation enzyme activity | |
| | | | | $^{\dagger}$2.5-40 | Lipid peroxidation; cellular damage; loss of redox homeostasis | |
| | | | $Dy^{3+}$ | $^{\dagger}$5-40 | Increased metabolic activity | Freitas et al. (2020b) |
| | | | | $^{\dagger}$2.5-40 | Increased tissue concentration | |
| | | | | $^{\dagger}$20-40 | Stimulation of antioxidant and biotransformation enzymes | |
| | | | | $^{\dagger}$2.5-40 | Lipid peroxidation; cellular damages; loss of redox homeostasis | |
| | | | $Gd^{3+}$ | $^{\dagger}$30-120 | Decrease in metabolic activity; lipid peroxidation; cellular damages; neurotoxicity | Henriques et al. (2019) |
| | | | | $^{\dagger}$30-60 | Stimulation of antioxidant enzymes | |
| | | | $LaCl_3 \bullet 7H_2O$ | $^{\dagger}10^2$-$10^5$ | Decrease in metabolic activity; increased activity of antioxidant enzymes; neurotoxicity; histopathological effects | Pinto et al. (2019) |
| | | adult | $LaCl_3$; $YCl_3$ | $^{\dagger}10^2$-$10^5$ | Abnormal development (La > Y) | Mestre et al. (2019) |
| | *Crassostrea gigas* | embryo | $La_2O_3$; $Y_2O_3$ | $^{\dagger}EC_{50}$ (24 h) La = 6.7; Y = 147.1<br>$^{\dagger}EC_{50}$ (48 h) La = 36.1; Y = 221.9 | Abnormal development (La > Y) | Moreira et al. (2020) |
| **Echinoderms** | *Paracentrotus lividus; Heliocidaris turbercolata; Arbacia lixula; Centrostephanus rodgersii* | embryo | $Gd(CH_3CO_2)_3 \bullet 4H_2O$ | $^{\phi}EC_{50=}$0.056–132 | Abnormal skeletogenesis | Martino et al. (2017a) |
| | | | $Gd(CH_3CO_2)_3 \bullet 4H_2O$ | $^{\phi}$20 (*P. lividus*) | Abnormal skeletogenesis; autophagy | Martino et al. (2017b) |
| | | | $Gd(CH_3CO_2)_3 \bullet 4H_2O$ | $^{\phi}$20 (*P. lividus*); $^{\phi}$0.5 and 5 (*H. tubercolata*) | Increased tissue concentration; decrease in $Ca^{2+}$ content; abnormal skeletogenesis; mis-regulation skeletogenesis genes | Martino et al. (2018) |
| | | embryo; sperm | $La^{3+}$; $Ce^{4+}$ | $^{\phi}EC_{50}$ La = 6.0 × $10^{-3}$ Ce = 1.9 × $10^{-3}$; | Decrease in mitotic activity, abnormal development (Ce); inhibition of fertilisation rate (10 $^{-5}$ M La and Ce); malformation in offspring from Ce-exposed sperm. | Oral et al. (2010) |
| | | | $LaCl_3$, $GdCl_3$; $YCl_3$ | $^{\phi}EC_{50}$ La = 6.6 × $10^{-4}$; Gd = 1.97 × $10^{-4}$; Y = 7.98 × $10^{-4}$ | Abnormal development | Pagano et al. (2015) |
| | | | $LaCl_3$; $CeCl_3$; $SmCl_3$; $GdCl_3$ | $^{\phi}10^{-3}$-$10^{-1}$ | Inhibition of mitotic activity (Sm, Ce > La, Gd) | |
| | | | $LaCl_3$; $CeCl_3$; $NdCl_3$; $SmCl_3$; $EuCl_3$; $GdCl_3$; $YCl_3$ | $^{\phi}10^{-1}$ | Inhibition of fertilisation success (Eu, Y > others) | |
| | | | | $^{\phi}10^{-2}$-$10^{-1}$ | Abnormal development in offspring from exposed sperm (La, Y> others); inhibition of mitotic activity (Ce > La; Sm) | |
| | | | $CeCl_3$, $SmCl_3$; $YCl_3$ | $^{\phi}10^{-3}$ | Increased levels of reactive oxygen species (ROS) | |
| | | | $CeCl_3$; $GdCl_3$ | $^{\phi}10^{-3}$ | Oxidative stress; lipid peroxidation | |
| | | | $LaCl_3$, $CeCl_3$, $GdCl_3$; $YCl_3$ | $^{\phi}10^{-3}$ | Increased levels of nitrites (NO) (Y > La > Gd > Ce)t | |
| | | | $LaCl_3$, $CeCl_3$, $NdCl_3$ $EuCl_3$, $SmCl_3$, $GdCl_3$, $YCl_3$ | $^{\phi}10^{-1}$ -10 | Abnormal development (La, Ce, Nd, Eu, Sm); mortality (Gd, Y at highest concentrations) | Trifuoggi et al. (2017) |
| | | | | $^{\phi}1 \cdot 10^2$ | Mitotic aberrations in interphase embryos | |
| | | | | $^{\phi}$10-$10^2$ | Abnormal development in offspring from exposed sperm | |
| **Fish** | *Anguilla anguilla* | adult | $LaCl_3 \bullet H_2O$ | $^{\dagger}$0.12 | Increased concentration in skinless body; increased catalase (CAT) activity; increased AchE activity | Figueiredo et al. (2018) |
| | | | $LaCl_3$ | $^{\dagger}$1.5 | | |

(continued on next page)

**Table 4** (*continued*)

| Cohort | Species | Life stage | Tested REY compound | Effect concentration (EC) | Effects | Reference |
|---|---|---|---|---|---|---|
| | | | | | Increased concentration (viscera > body > head);<br>DNA damages; lipid peroxidation; expressional suppression of heat shock proteins | Figueiredo et al. (2020) |
| | *Takifugu niphobles* | sperm | $GdCl_3$ | $^{\phi}$10-40 | Inhibition of osmosis-initiated mobility | Krasznai et al. (2003) |

external $CaCO_3$ structures (marine biogenic calcification) such as exoskeletons and shells. Ponnurangam and co-authors studied accumulation of REY in blue mussel (*Mytilus edulis*) shells and showed that free $REY^{3+}$ are taken up and incorporated into the shells, with middle-REY being taken up more efficiently than LREY and HREY (Bau et al., 2010; Ponnurangam et al., 2016). The authors further detected distinct REY signatures, such as small positive $Y_{SN}$ and $Gd_{SN}$ anomalies, in the mussel shells reflecting REY patterns of the seawater they grew in, making them a suitable species for REY biomonitoring. Pérez de Nanclares and colleagues (2014) observed increased incorporation of 4 REY (Ce, Nd, Pr and Dy) in Atlantic salmon (*Salmo salar*) scales when exposed to REY-enriched feed and a decreasing in REY concentration over time at the end of the exposure, as effect of continuous $Ca^{2+}$ deposition diluting the previously deposited REY. The same authors highlighted the low digestibility of REY compounds, suggesting that deposition of REY in fish scales is not an effect of absorption in the gastro-intestinal tract but rather a result of direct absorption from seawater.

Studies analysing organ/tissue distribution of REY in marine organisms are scarce, with mostly only the edible parts of the organisms, such as fish muscle and whole soft-tissue of shellfish and crustaceans being analysed (Table 3). The few studies conducting tissue-specific comparative analyses of REY in marine vertebrates (fish, seals, birds), report higher concentrations in internal/detoxification organs, such as the liver and kidneys, compared to muscle tissue, with the latter sometimes featuring REY concentrations below detection limits (MacMillan et al., 2017; Squadrone et al., 2020). Further, the hypothesis that REY follow/replace $Ca^{2+}$ in organisms (Bau et al., 2010; Leonard et al., 2019; Ponnurangam et al., 2016; Reindl et al., 2021; Squadrone et al., 2019a) is further supported by the fact that REY concentrations determined in fur, feathers, shells and exoskeletons largely exceed those in soft tissue and organs (Akagi and Edanami, 2017; Squadrone et al., 2019b; Reindl et al., 2021) (Table 1). This highlights the need to also analyse $Ca^{2+}$-incorporating tissues in addition to edible tissues and organs in REY biomonitoring studies as these structures could constitute useful proxies for the evaluation of spatial and temporal patterns of environmental presence and anthropogenic environmental enrichment of REY.

### 5.2. *Eco-toxicological effects*

Data on effects and toxicological mechanisms of REY in marine organisms is scarce and therefore not yet conclusive. Effects-studies consider concentrations which are several order of magnitude higher than the reported environmental levels (Tables 1, 2 and 4), further no literature exists on the ecological effects of these emerging contaminants at community and ecosystem levels.

According to our literature search criteria, only 17 research papers (from 2003 to 2020) specifically evaluated the effects of REY on marine organisms, of which 29% ($n = 5$) were from 2020. These studies evaluated different elements and species for physiological and toxicological endpoints, thus making it difficult to establish clear trends across studies.

Tai et al. (2010) investigated the effect of 13 lanthanides and Y (tested as REY salts) on the growth of the green unicellular algae *Skeletonema costatum*, reporting similar effect concentrations (median effective concentration - $EC_{50}$) of approximately 30 μmol $L^{-1}$ (~7–13 mg $L^{-1}$) for the lanthanide elements, and an $EC_{50}$ of 43 μmol $L^{-1}$ (~8 mg $L^{-1}$) for Y. Further, mixtures of the 13 lanthanide elements exhibited similar inhibitory effects on algae growth when the sum of concentrations were equal to the concentration of single elements, indicating a similar mode of action.

Differences in effect concentrations (48 h, immobilisation) between REYs were reported for the brine shrimp *Artemia salina*, with Nd ($EC_{50} =$ 47 mg $L^{-1}$) being more toxic than La (78 mg $L^{-1}$) and Sm (122 mg $L^{-1}$) with both synergistic ($La^{3+}$+$Nd^{3+}$; $Nd^{3+}$+$Sm^{3+}$) and antagonistic ($La^{3+}$+$Nd^{3+}$+$Sm^{3+}$) effects observed in co-exposures (Bergsten-Torralba et al., 2020).

Bivalves seem to be more sensitive compared to algae and crustaceans, with a higher toxicity of LREY compared to HREY reported in several studies. La was described as more toxic to oyster embryos (*Crassostrea gigas*) compared to the Y, with $EC_{50}$ concentrations (development) < 50 μg $L^{-1}$ for La, and approximately 150 μg $L^{-1}$ (24 h) and 220 μg $L^{-1}$ (48 h) for Y (Moreira et al., 2020). Similarly, La was found to be more toxic (development; $EC_{50}$ ~ 50 μg $L^{-1}$) compared to Y ($EC_{50}$ 800 μg $L^{-1}$) for developing Mediterrean mussel *Mytilus galloprovincialis* (Mestre et al. 2019).

Adverse and toxic effects of La, Nd, Gd, Dy were further described for adult Mediterrean mussel. In these studies (28 day exposure), REY exposure at concentrations above 2.5 μg $L^{-1}$ resulted, amongst others, in cellular damage, oxidative stress, metabolic changes, neurotoxic effects and loss of redox balance (Freitas et al., 2020a, b; Henriques et al., 2019; Pinto et al., 2019).

Interestingly, La appeared to exert effects at higher concentrations compared to Nd, Gd and Dy (Freitas et al., 2020a, b; Henriques et al., 2019; Pinto et al., 2019). Gd caused a significant inhibition of acetylcholinesterase (AchE) activity at concentrations above 15 μg $L^{-1}$, indicating neurotoxic activity, which could be related to $Gd^{3+}$ acting as $Ca^{2+}$ or calcium-gated ion channel blocker (Henriques et al., 2019; Palasz and Czekaj, 2000).

REY (Y, La, Ce, Nd, Sm, Eu and Gd) exposure was reported to impact the development (damaged skeletal differentiation and/or abnormal blastulae or gastrulae stages) and survival of sea urchin early life stages (*Paracentrotus lividus*, *Sphaerechinus granularis* and *Arbacia lixula*) in a concentration-related manner, with exposure concentrations reaching from 0.01 to 100 μM (Trifuoggi et al., 2017; Pagano et al., 2015). Developmental effects were also observed in offspring fertilised with sperm exposed to REY, although at higher exposure concentrations. Exposure to these REY was further shown to cause oxidative stress in *P. lividus* early life stages (Pagano et al., 2015). Differences in sensitivity between species were also reported in a study by Martino et al. (2017a), comparing effects of Gd on the development (skeleton formation) of two European (*P. lividus* and *A. lixula*) and two Australian sea-urchin species (*Heliocidaris tuberculata* and *Centrostephanus rodgersii*). With an $EC_{50}$ of 0.056 μM (tested and reported as Gadolinium (III) Acetate Tetrahydrate: 22.8 μg $L^{-1}$) *H. tuberculata* was by far (2–3 orders of magnitude) the most sensitive species. Effects of Gd on the developing skeleton (impairment of skeleton growth, asymmetrical spicule formation) were related to increased autophagic activity and impaired biomineralisation ($Ca^{2+}$ uptake) in developing embryos, with $Gd^{3+}$ acting as blocker/competitor for $Ca^{2+}$ at Ca ion channels (Martino et al., 2017 a,b; 2018; David et al., 1988). Further, gene expression of skeletogenic-related genes was altered following Gd exposure (Martino et al., 2018).

Effect studies on marine vertebrates are scarce. Figueiredo et al. (2018, 2020) observed uptake of La in glass eels (*Anguilla anguilla)* at exposure concentrations of 0.12 and 1.5 μg $L^{-1}$, which was accompanied by increased DNA damage, lipid peroxidation and a suppression of heat shock proteins. Accumulation and effects were more pronounced at increased exposure temperature, highlighting the role of changing climatic conditions in affecting the toxicity of contaminants (Figueiredo et al., 2020). La exposure also caused a significant increase in the activity of AchE in glass eels exposed to 0.12 μg $L^{-1}$, which was, however, not observed at 1.5 μg $L^{-1}$ (Figueiredo et al., 2018, 2020). Gadolinium (10–40 μM $GdCl_3$; ~2.6–10.5 mg $L^{-1}$) was further shown to (reversibly) inhibit the osmosis -initiated motility of puffer fish (*Takifugu niphobles*) sperm in a dose- and incubation time-dependent manner (Krasznai et al., 2003). Inhibitory effects were described to be related to Gd interference with $Ca^{2+}$ fluxes, intracellular $Ca^{2+}$ mobilisation, and alterations of the isoelectric point in motility-related proteins (Krasznai et al., 2003). The inhibitory effect of Gd on cell motility, functioning as stretch activated channel blocker, has also previously been described in other vertebrate cells (e.g. Munevar et al., 2004).

## 6. Knowledge gaps and perspectives

Research on REY as contaminants of emerging concern is still in its infancy and thus considerable knowledge gaps exist especially from an ecotoxicological perspective. The key knowledge gaps as well as advice for future research topics are summarised below in four thematic areas: 1) Harmonisation of reporting data and units; 2) ACC-REY environmental behaviour and fate; 3) Toxicological effects on organisms; 4) Ecological implication and risk assessment of different REY with respect to individual bioaccumulation levels and effects at community and ecosystem level.

### *6.1. Harmonisation of reporting data and units*

Particular attention should be given to harmonising the way in which data are reported to promote comparability between studies and data. There is currently a clear distinction between the geochemical and the biological research fields on how REY concentration data are presented. While normalized concentration patterns of individual REY (e.g. for seawater and sediments) are reported within geochemical research, most biological studies treat REY either as a single contaminants or as homologue group, only reporting single or total REY concentrations without distinguishing between the different elements. Furthermore, biological studies do frequently not account for environmental background levels.

However, for studies aiming to evaluate the potential risks associated with anthropogenic release of REY, it is essential to compare and report REY as normalized patterns and compare with geogenic background data whenever possible, as anthropogenic REY enrichment is identifiable from peaks (anomalies) in otherwise smooth $REY_{SN}$ patterns.

Further, a distinction between different REY is recommendable, as some elements can represent a greater concern due to (i) being released from anthropogenic sources at higher levels, (ii) being more environmentally stable or bioavailable, or (iii) eliciting different toxicological effects.

### *6.2. Marine environmental behaviour and fate*

The environmental behaviour and fate of chemical contaminants in the marine environment can significantly affect their potential for exposure to organisms and subsequent likelihood for causing toxicological effects. Well documented exposure data and a comprehensive understanding of REY behaviour and fate in the marine environment is therefore essential for the development of risk assessment and risk mitigation strategies. Consequently, understanding the physical-chemical and biological processes that affect REY fate in the environment is critical.

While the geochemical processes affecting geogenic REY mobilisation from natural sources to the water column and sediments are relatively well-established, there is a lack of research evaluating the mechanism of transport and chemical behaviour (e.g. dissolution, aggregation, fractionation and bioaccumulation) of ACC-REY.

As ACC-REY could be potentially more chemically stable and soluble in the marine environment than geogenic REY, the extent of such contamination needs be evaluated and quantified, including the assessment of their stability, persistence and bioavailability for organisms in the marine environment.

### *6.3. Organismal uptake and toxicological effects*

Data on the toxicological effects of REY on marine organisms are generally scarce. Although adverse effects such as abnormal development, cellular and tissue damage, oxidative stress, and neurotoxicity and have been reported in marine organisms following exposure to individual REY, more data is needed to identify target species and tissues (with different functions) prone to accumulate REY as well as specific mechanisms of toxic action, dose-dependent effects, and effect threshold concentrations. Furthermore, assessments comparing the effects of ACC-REY with naturally occurring geogenic forms of REY, also accounting for environmentally relevant exposure concentrations which at the current status are largely neglected, are required to determine the risks associated with anthropogenic REY releases.

### *6.4. Ecological implications and risk*

Evaluations of the ecological implications of REY contamination are, to date, very scarce for aquatic environments and almost entirely lacking for the marine environment. To determine if there is any ecological risk associated with anthropogenic REY contamination, it is suggested that the following aspects warrant further research: 1) comprehensive assessment of REY in field collected organisms characterised by various ecological traits (e.g. feeding modes; life habits; trophic position); 2) targeted laboratory and mesocosm trophic transfer studies to determine (or exclude) the potential for REY to be accumulated (or alternatively egested) and transferred through marine food webs; 3) generation of quantitative data providing information on the impacts of REY on natural populations (e.g. variation in size, increased mortality and decreased reproductive output) and communities (e.g. loss or changes in abundances of associated species), as well as on the risk of trophic cascade effects; 4) evaluation of the impacts of REY and ACC-REY at environmentally relevant concentrations and conditions for example when combined with other global change-related natural and anthropogenic stressors.

## 7. Conclusion and final remarks

In this review we examined and discussed the current knowledge regarding the sources, distribution and effects of REY in the marine environment, highlighting knowledge gaps and suggesting further research needs to assess the impact of REY at organismal, community and ecosystem levels. As research progresses, there is clear need for harmonisation across geochemical and biological studies in terms of data treatment and reporting, so that a more complete picture on the potential threat of REY to organisms and ecosystems emerges. It is essential to establish an improved understanding of the factors affecting the transport of REY from anthropogenic sources to the marine environment, for which sufficient empirical data are currently lacking. Finally, deepening knowledge of the persistence, bioavailability and effects of geogenic *vs* ACC-REY across species, communities and ecosystems will be critical to assess any potential environmental risk related to these emerging contaminants and for assisting in the development of future regulatory frameworks.

## Funding

This work has been conducted under the framework of the project ELEMENTARY and funding has been provided by the Norwegian Research Council (grant agreement No. 301236).

## Credit author statement

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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.

## Acknowledgement

We wish to express our gratitude to Prof. Michael Bau for the theoretical support and quality check of the geochemical part of this review.

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