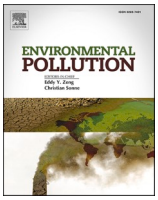
Environmental Pollution 335 (2023) 122300

Contents lists available at ScienceDirect

Environmental Pollution

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



Biomonitoring of rare earth elements in Southern Norway: Distribution, fractionation, and accumulation patterns in the marine bivalves *Mytilus* spp. and *Tapes* spp.☆

Lyen Castro [a], Julia Farkas [b,*], Bjørn Munro Jenssen [a,c], Stefania Piarulli [b], Tomasz Maciej Ciesielski [a,c]

[a] Department of Biology, Norwegian University of Science and Technology, 7491, Trondheim, Norway
[b] SINTEF Ocean, Climate and Environment, 7465, Trondheim, Norway
[c] Department of Arctic Technology, The University Center in Svalbard, 9171, Longyearbyen, Norway

ARTICLE INFO

Keywords:
Rare earth elements
Gadolinium-based contrast agent
Gadolinium anomaly
Mussel
Bivalves
Biomonitoring

ABSTRACT

Growing extraction and usage of rare earth elements and yttrium (REY) for medical and industrial applications has resulted in increased discharges into the marine environment. Using *Mytilus* spp. Mussels and *Tapes* spp. clams as bioindicator organisms, we analyzed 15 REY in soft tissues of specimens collected at two potentially polluted sites in Southern Norway: in the vicinity of an industry producing gadolinium-based MRI contrast agents (GBCAs) (Lindesnes) and in an industrially-affected fjord (Porsgrunn). The spatial distribution of REY and shale-normalized fractionation patterns were determined to assess the potential anthropogenic contribution of REY at the sites. At both sites, the REY fractionation pattern in soft tissue was characterized by enrichment of light rare earth elements (LREE) over heavy rare earth elements (HREE), while also displaying negative cerium and small positive gadolinium (Gd) anomalies. LREEs contributed to over 80% of the total REY concentrations, with increasing relative enrichment following higher total REY. Gd anomalies remained conserved in most sites despite significant differences in total REY; however, a high Gd anomaly ($Gd/Gd^* = 4.4$) was found downstream of the GBCA industry spillwater outlet, indicating biotic uptake of excess anthropogenic Gd at this site. Total REY concentrations in clams in Porsgrunn were one order of magnitude higher than in mussels in Lindesnes. This may be attributable to freshwater influences in Porsgrunn, where clams collected closer to the river mouth had significantly higher total REY concentrations. This study constitutes the first assessment of REY concentrations in marine bivalves in Norway and can provide useful information for future biomonitoring studies on REY contamination.

1. Introduction

The use of rare earth elements (REEs) and yttrium (Y), which has similar properties to REE, has grown in terms of importance and diversity of industrial applications, reaching from medicine to advanced technology, leading to their growing demand and extraction. Among their different applications, REEs are used in catalysis, glassmaking, metallurgy, agriculture, battery alloys, ceramics and permanent magnets. Due to their catalytic, magnetic, and electronic properties, REEs have become crucial to technological advances, allowing devices to perform at reduced energy consumption and greater efficiency (Castor & Hedrick, 2006). Owing to their economic importance and supply risk, they currently belong to the list of Critical Raw Materials as defined by the European Union (Nuss & Blengini, 2018).

The waste produced by the urban, industrial, agricultural, or medical use of these critical metals, i.e. REE and Y (abbreviated REY), follows several direct or indirect release pathways (Piarulli et al., 2021), many of which can eventually reach the marine environment, the ultimate sink for these contaminants. This increasing release into the environment has led to REY being considered as contaminants of emerging concern (Gwenzi et al., 2018).

REY comprise 15 lanthanides (La–Lu) and yttrium (Y) and are a

☆ This paper has been recommended for acceptance by Christian Sonne.

* Corresponding author. SINTEF Ocean, Brattørkaia 17C, 7010, Trondheim, Norway.
E-mail address: julia.farkas@sintef.no (J. Farkas).

https://doi.org/10.1016/j.envpol.2023.122300
Received 19 April 2023; Received in revised form 10 July 2023; Accepted 31 July 2023
Available online 1 August 2023

relatively chemically uniform group of elements. Although there has been no worldwide consensus in the literature regarding their classification, REY are commonly divided by electron configuration into light rare earth elements (LREE) and heavy rare earth elements (HREE). LREE consist of the elements from lanthanum (La) to europium (Eu), while the HREEs include the elements from gadolinium (Gd) to lutetium (Lu), and yttrium (Gonzalez et al., 2014). This coherent group of elements has several shared chemical properties, such as having the same configuration of valence electrons in the outer shell. As their 4f orbitals are progressively filled, unpaired electrons are responsible for giving all lanthanides (except La and Lu) strong magnetic properties. REY also share a common trivalent oxidation state, except cerium (Ce) and Eu which have additional oxidation states. This, along with their decreasing ionic radii with increasing atomic number, allows them to replace elements with similar ionic radii, and act coherently in geochemical processes and as geochemical tracers (Zepf, 2013; Migaszewski & Gałuszka, 2015). With the exception of promethium (Pm), which has no long-lived isotopes, REY are not rare in the environment, the more abundant rare earths (e.g. La, Ce) have similar crustal concentrations to industrial metals such as copper, lead, zinc, and chromium (Haxel, 2002).

Often, the abundance of each REY is expressed relative to its concentration in shale (REY_{SN}) or chondritic meteorites and presented in a logarithmic scale plotted as a function of atomic number, known as the "Masuda-Coryell plot" (Coryell et al., 1963). This normalization to a natural reference system allows to easily discern and quantify deviations (called anomalies) in individual REY abundance from the smooth trend, and facilitates better comparison of REY patterns across different materials. The distinct REY distribution patterns that occur in different environmental compartments can serve as potential tracers to discriminate between geogenic and anthropogenic sources (Migaszewski & Gałuszka, 2015).

Gadolinium is one of the most commercially exploited REY due to its large use in medical diagnostics. It is typically chelated with a linear or macrocyclic ligand to form gadolinium-based contrast agents (GBCAs) to enhance the quality of magnetic resonance imaging (Trapasso et al., 2021). Because GBCAs are not removed from the water phase during wastewater treatment, elevated Gd concentrations have been reported globally in surface waters near populated areas, particularly near hospital effluents and later in the effluent of wastewater treatment plants (Lerat-Hardy et al., 2019; Le Goff et al., 2019). In rivers draining densely industrialized areas in Central Europe and North America, shale-normalized rare earth element patterns are characterized by pronounced positive Gd anomalies (Bau & Dulski, 1996) compared to less populated, non-industrialized areas, which do not show such anomalies. Positive Gd anomalies in aquatic environments are thus of concern for the aquatic organisms that can be constantly exposed to these contaminated waters.

Filter-feeding benthic organisms such as bivalve molluscs are often used as bioindicators of contamination due to their sessile lifestyle, widespread distribution, high bioaccumulation capacity of a wide range of contaminants, and constant exposure to potentially contaminated organic and inorganic particles (Beyer et al., 2017). Concentrations in the soft tissue of bivalves can reflect a temporally integrated measure of bioavailable contamination or pollution in a given area. The soft tissue of bivalves has also been reported to accumulate up to tenfold higher concentrations of REY compared to the shell (Akagi & Edanami, 2017). There has been a growing number of recent studies around the world that have investigated the accumulation and fractionation patterns of REY in the soft tissues of various marine and freshwater bivalves near metropolitan sites, including *Mytilus edulis, Mytilus galloprovincialis, Crassostrea gigas, Ruditapes philippinarum,* and *Corbicula fluminea* (Briant et al., 2021; Figueiredo et al., 2022; Rodríguez-Velarte et al., 2022; Akagi & Edanami, 2017; Wang et al., 2019; Zhao et al., 2022; Pereto et al., 2020). Thus, bivalves appear to be a suitable bioindicator for REY contamination in marine ecosystems.

In this study, we assessed the accumulation of REY in the soft tissues of marine bivalves from two industrially-affected sites in Southern Norway. We measured individual and total REY concentrations in *Mytilus* spp. Mussels within the vicinity of a GBCA industry in Lindesnes, and in *Tapes* spp. clams near a heavily industrialized area in Porsgrunn. The specific objectives of this study were to (1) investigate spatial variations in REY accumulation among the different sampling sites against background values, (2) characterize the shale-normalized REY distribution patterns in each species, and from this (3) quantify Gd anomalies at the two sites to identify potential anthropogenic Gd accumulation in bivalve soft tissue.

2. Material and methods

2.1. Study sites and sampling

Field sampling was conducted in the south of Norway in May and June 2021. The mussels *Mytilus* spp. were handpicked at four sites in Lindesnes (LE1, LE2, LE3, and LE4) (Fig. 1). Sites LE2 through LE4 were taken in proximity to the offshore pipe outlet (58° 1′ 22″ N, 7° 7′ 3″ E) of a GBCA manufacturer. The reference site mussels, LE1, were taken in Njervesfjorden, 3 km away northeast (58° 2′ 25″ N, 7° 9′ 17″ E). LE2 and LE3 mussels were taken north of the pipe, the former from a rocky beach (58° 1′ 39″ N, 7° 7′ 10″ E) located inside the shallow Ramslandsvågen bay and the latter from a small islet (58° 1′ 26″ N, 7° 7′ 5″ E) near the outlet of the pipe. LE4 mussels were collected from a rocky beach (58° 1′ 12″ N, 7° 6′ 47″ E) located southwest of the pipe.

The clams *Tapes* spp. were collected from three sites (PO1, PO2, and PO3) along the Frier fjord in Porsgrunn, an important industrial area in Norway that houses petrochemical industries and processing facilities for fertilizer, porcelain, polyvinyl chloride, and several metals. The main sewage from the Skien and Porsgrunn municipalities is treated in the Knardalstrand wastewater treatment plant (WWTP), which discharges at the bottom of the Skien River and into Frier fjord (Fig. 1) (Staalstrøm, 2013). Site PO2 (59° 7′ 8″ N, 9° 35′ 30″ E) is located west of the discharge point of the Knardalstrand WWTP (59° 07′ 18″ N, 9° 36' 9" E), in the direction of the outflow of the Skien river. PO3 clams were taken from the coast of the Herøya industrial park peninsula (59° 6′ 40″ N, 9° 38′ 18″ E), close to the outlet of the enclosed fjord Gunnekleivfjorden. The reference site, PO1, was located at the southern far end of Frier fjord (59° 2′ 53″ N, 9° 39′ 36″ E).

The mussels and clams were packed and frozen at −20 °C within a few hours after collection, and kept frozen until further sample preparation and analyses.

2.2. Tissue sample preparation

The frozen bivalve specimens were thawed to room temperature and the shells were cleaned with ultrapure water (Q-option, Elga Labwater, Veolia Water Systems LTD, UK) to remove debris. Each individual was weighed on an electronic top-loading balance (Sartorius BP 4100) and shell length was measured along the anterior-posterior axis of the bivalve using the digital caliper to the nearest 0.01 mm. The whole soft tissue was separated from the shell using a titanium knife and collected into polypropylene vials. Specimens were sorted by shell length and smaller individuals were pooled to obtain sufficient dry weight (100–500 mg) for elemental analysis. The total number of bivalves collected per site were: 50 (LE1), 26 (LE2), 100 (LE3), 95 (LE4), 16 (PO1), 38 (PO2), and 16 (PO3). After pooling smaller individuals of similar size, the resulting number of samples per each site were: 4 (LE1), 15 (LE2), 22 (LE3), 8 (LE4), 5 (PO1), 12 (PO2), and 5 (PO3). Biometric data and sample pooling information can be found in Supplementary Tables S1 and S2. All collected soft tissues were freeze-dried for a period of 72 h.

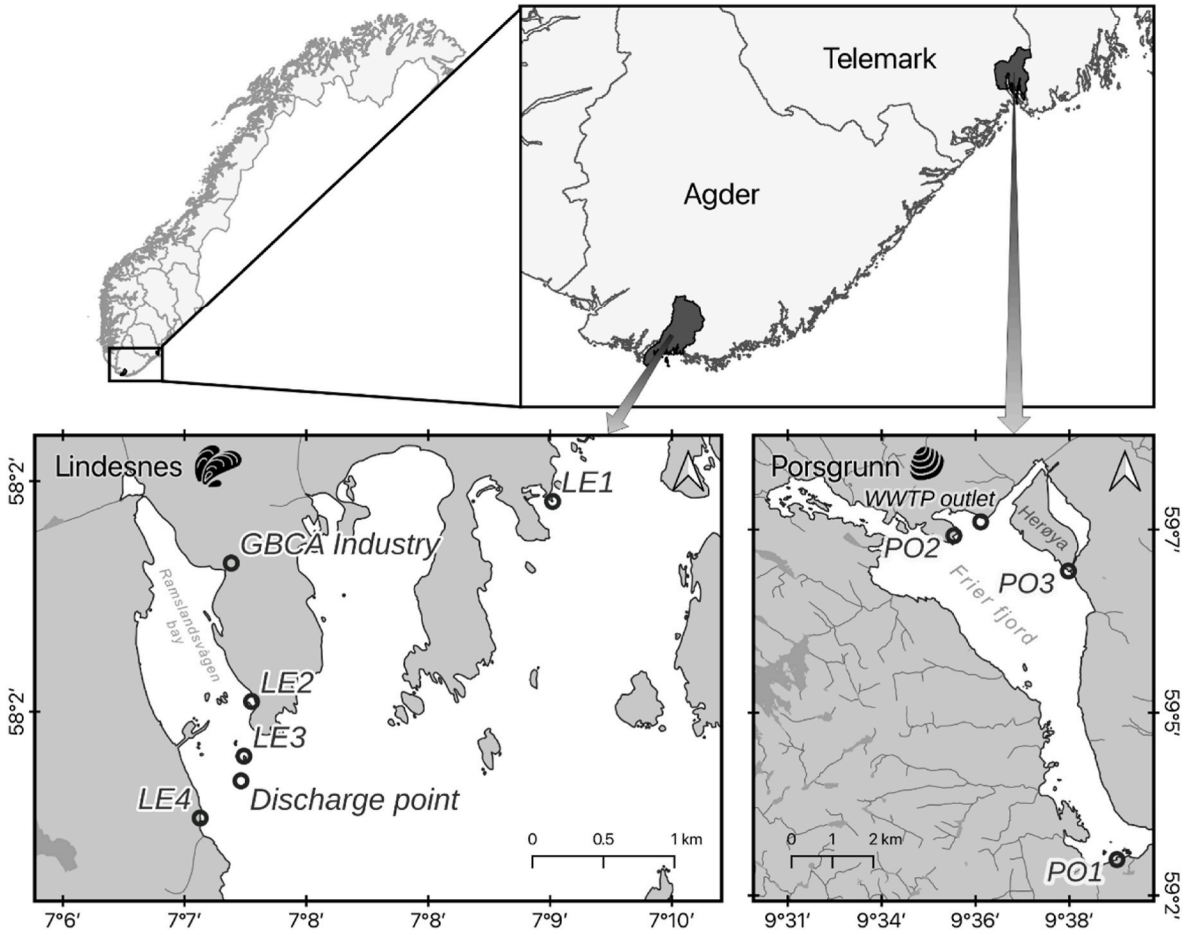


Fig. 1. Locations of the bivalve sampling sites around the cities of Lindesnes and Porsgrunn, Norway during the field sampling campaign in May to June 2021. LE1 and PO1 are the reference sites for Lindesnes and Porsgrunn, respectively. Geographical mapping was performed using QGIS 3.26.3 (data source: Global Administrative Areas database and the Norwegian Water Resources and Energy Directorate).

2.3. Elemental analysis

For multi-elemental analysis, 100–500 mg of freeze-dried tissue was weighed into a clean polytetrafluoroethylene vial and 5 mL nitric acid (50% v/v HNO_3, ultrapure grade, purified from HNO_3 [AnalaR NORMAPUR®, VWR] in a sub-boiling distillation system, Milestone, SubPur, Sorisole, BG, Italy) was added. Sample digestion was performed in a Milestone ultraCLAVE microwave reactor system (1000 W, 160.0 bar) with a gradual, step-by-step temperature ramp from 50 to 245 °C in 80 min. Each digested sample was then quantitively transferred to a 50 mL metal-free polypropylene tube and diluted with ultrapure water to 51 g. The final weight of each solution was recorded on an electronic top-loading balance (Sartorius BL210S) to the nearest 0.01 g. Three procedural blanks and three replicates of certified reference material (CRM No. 668 mussel soft tissue, Community Bureau of Reference, European Commission) were digested together in each sample digestion batch (results can be seen in Table S3). Finally, the samples were stored at room temperature until analysis.

Concentrations of 15 REY (La, Ce, Pr, neodymium [Nd], samarium [Sm], Eu, Gd, terbium [Tb], dysprosium [Dy], holmium [Ho], erbium [Er], thulium [Tm], ytterbium [Yb], Lu, including Y) were determined via using a triple quadrupole inductively coupled plasma mass spectrometry instrument (Agilent 8900 ICPQQQ, Agilent Technologies, USA) with ^{103}Rh, ^{185}Re and ^{193}Ir as internal standards (Inorganic Ventures, USA). The limits of quantification (LOQ) were set to 3 times the instrumental detection limits (IDL) adjusted to the sample amount used for analysis. The IDLs were estimated from the subsequent analysis of solutions, containing decreasing, low concentrations of the element. Finally, the concentration with a signal that is clearly distinguishable from the background noise level and resulting in a relative standard deviation of approx. 20% was selected as IDL.

All elemental concentrations in soft tissues were found to be above the LOQ, with the exception of Tm (n below LOQ = 13) and Lu (n below LOQ = 17) for mussel samples (n = 49). Values below LOQ were substituted with the instrumental LOQs during statistical analysis and graphing.

REY concentrations are all presented in micrograms per gram soft tissue dry weight (μg g^{-1}, d.w.). In order to determine the REY fractionation pattern relative to the continental source, shale-normalized (REY_{SN}) plots were made by dividing REY abundances by their corresponding values in European shale (Bau et al., 2018). This normalization to shale eliminates the 'saw-tooth' Oddo-Harkins effect in the natural abundance of REY in which elements with an odd atomic number are less abundant than adjacent even-numbered elements. Using the calculated REY_{SN} values, Ce anomalies (Ce/Ce*) and Gd anomalies (Gd/Gd*) were quantified using the equations described in Ponnurangam et al. (2016). The LREE enrichment was determined using the ratio of La_{SN} to Yb_{SN} ($[La/Yb]_{SN}$) (Briant et al., 2021). All data in the text were presented as their mean ± standard deviation.

2.4. Statistical analysis

Prior to analysis, the Shapiro-Wilk test was used to check the normality of the underlying residuals of each variable. This was

followed by Levene's test on the original data to determine if the samples met the assumption of homoscedasticity for a parametric test. To detect significant differences in REY concentration or biological parameters among sites, the omnibus One-Way ANOVA test was used, followed by Tukey-Kramer *post hoc* test for pairwise comparisons. For data with unequal variance (*i.e.* ΣREY, ΣHREE, ΣLREE), Welch's ANOVA with the Games-Howell *post hoc* test was utilized. Due to the non-normal distribution of the data, Gd anomalies per site were compared using the nonparametric Kruskal-Wallis test followed by a Dunn-Bonferroni *post hoc* test. All analyses were performed using SPSS (Version 29) (IBM Corp, 2021) at a 95% confidence level.

3. Results

3.1. Size and body weight distribution of the collected bivalves

The mussels collected at Lindesnes had variable shell lengths, ranging between 13 and 34 mm. The mussels at LE1 (the reference site) (19 ± 12 mm) were comparable in size to all other sites in Lindesnes ($p > 0.222$). On the other hand, mussels from sites LE2 (34 ± 7 mm) and LE3 (28 ± 8 mm) were of comparable shell lengths ($p = 0.072$), but significantly different from the smaller LE4 mussels (13 ± 2 mm) ($p < 0.001$). *Tapes* spp. from all three sites in Porsgrunn were uniform in terms of shell length (29–31 mm, $F_{2,19} = 0.75$, $p = 0.486$).

3.2. Total REY concentrations

The concentrations of all REY in soft tissues per site are reported in Table 1. Total REY concentrations (ΣREY) in Lindesnes mussels ranged from 0.95 to 4.88 μg g^{-1}. The site LE2 located at the mouth of the enclosed bay had significant, fourfold higher ΣREY than the other sites in Lindesnes ($F_{3,14.56} = 43.20$, $p < 0.001$). The rest—the northern site LE3 and southwestern site LE4 had comparable ΣREY levels with the reference site ($p = 0.615$ and $p = 0.729$, respectively). Total LREE concentrations (ΣLREE) ranged from 0.76 to 4.2 μg g^{-1} and the spatial distribution among the sites followed the same pattern as ΣREY (Fig. 2A). In terms of total HREE concentrations (ΣHREE), LE2 remained the highest, but this time site LE4 had become significantly different from the reference and LE3 ($F_{3,14.64} = 36.09$, $p < 0.001$) due to an increased contribution of Gd, despite it retaining similar total REY concentrations.

Tapes spp. collected in Porsgrunn had accumulated high concentrations of ΣREY in its soft tissue, from 45 ± 12 μg g^{-1} at the reference site (PO1), reaching up to 72 ± 22 μg g^{-1} in PO2, and 64 ± 15 μg g^{-1} in PO3 (Fig. 2B). For both ΣREY and ΣLREE concentrations, the trends were decreasing from the river mouth to the southern end of the fjord (PO2 > PO3 > PO1). The clams at PO2 had significantly higher ΣREY ($p = 0.018$) than at the reference site (PO1). Midway at PO3 (Herøya peninsula), ΣREY levels were comparable to both the reference site ($p = 0.143$) and PO2 ($p = 0.669$). ΣHREE concentrations did not differ between the clams at all three sites ($p = 0.090$).

3.3. REY_{SN} patterns and anomalies

Shale-normalized REY values in the analyzed soft tissues were calculated (Supplementary Table S4), and the overall fractionation patterns in bivalves from Lindesnes and Porsgrunn are displayed in Fig. 3. The general shape of the REY_{SN} curves of both *Mytilus* spp. and *Tapes* spp. is marked by a decline from LREE to HREE, a positive Gd anomaly, and a negative Ce anomaly.

In Lindesnes, LREE enrichment, represented by $[La/Yb]_{SN}$, was most pronounced in site LE2, in which the ratio (4.97 ± 1.54) was more than double compared to the rest of the locations (Supplementary Table S4). LE4 had the lowest ratio, as reflected by its flatter pattern (Fig. 3A), which was affected by one mussel with exceptionally high HREE enrichment. In Porsgrunn, clams exhibited very high LREE enrichment ($[La/Yb]_{SN} = 12.4–15.8$), over twice that of Lindesnes mussels.

The fractionation patterns in both bivalves in this study featured Ce depletion and Gd enrichment relative to other REEs. Porsgrunn clams showed greater Ce depletion, indicated by having a more negative Ce anomaly (Ce/Ce* = 0.70–0.76), compared to Lindesnes mussels (Ce/Ce* = 0.84–0.97). In terms of Gd enrichment, the positive Gd anomalies exhibited by both mussels and clams in southern Norway were all small (Gd/Gd* = 1.24–1.41), with the exception of mussels in site LE4 (Gd/Gd* = 4.43 ± 1.30). In Porsgrunn, the level of Gd enrichment was comparable for all sites ($H(2) = 4.887$, $p = 0.087$). In Lindesnes, the Gd anomalies were comparable between LE2 (Ramslandvågen bay) and the reference, whereas LE3 (north islet) displayed a significantly higher anomaly compared to both. There was a clear significant difference, however, between these three sites and LE4 (southwestern site) ($H(3) = 29.184$, $p < 0.001$), as manifested in the very pronounced Gd peak shown in Fig. 3A. This threefold higher, significantly large anomaly indicates a source of excess Gd in the environment affecting site LE4.

Table 1
Mean REY concentrations (μg g^{-1} dry weight) in the soft tissues of *Mytilus* spp. and *Tapes* spp. collected during May–June 2021 in southern Norway.

Element	*Mytilus* spp.								*Tapes* spp.						*LOQ (ng mL^{-1})*
	LE1 ($n = 4$)		LE2 ($n = 15$)		LE3 ($n = 22$)		LE4 ($n = 8$)		PO1 ($n = 5$)		PO2 ($n = 12$)		PO3 ($n = 5$)		
	mean	sd	mean	sd	mean	sd	mean	sd	mean	sd	mean	sd	mean	sd	
La	0.23	0.026	1.5	0.69	0.27	0.091	0.22	0.026	18	6.1	30	11	27	6.4	0.005
Ce	0.3	0.032	1.8	0.98	0.35	0.078	0.3	0.035	13	3.4	20	6.3	18	4.2	0.005
Pr	0.044	0.0053	0.2	0.091	0.048	0.012	0.042	0.0048	1.9	0.54	3.1	0.98	2.8	0.67	0.005
Nd	0.17	0.024	0.66	0.31	0.18	0.05	0.16	0.018	6.5	1.6	10	3	9	2.2	0.005
Sm	0.03	0.0045	0.096	0.045	0.033	0.0085	0.03	0.003	0.92	0.19	1.3	0.31	1.2	0.26	0.005
Eu	0.0052	0.00048	0.015	0.0073	0.0056	0.0015	0.0056	0.00091	0.13	0.028	0.17	0.037	0.15	0.034	0.005
Gd	0.028	0.0047	0.096	0.046	0.033	0.0089	0.094	0.026	0.82	0.17	1.2	0.26	1	0.22	0.005
Tb	0.0028	0.00045	0.0097	0.0047	0.0031	0.00075	0.0028	0.00023	0.091	0.028	0.11	0.026	0.1	0.021	0.001
Dy	0.019	0.002	0.063	0.03	0.02	0.0044	0.021	0.0029	0.44	0.19	0.57	0.13	0.47	0.089	0.005
Ho	0.0033	0.00037	0.011	0.0054	0.0037	0.00079	0.004	0.0011	0.08	0.04	0.098	0.026	0.08	0.014	0.005
Er	0.0091	0.0012	0.03	0.013	0.0098	0.0019	0.011	0.0045	0.21	0.13	0.24	0.084	0.2	0.031	0.005
Tm	0.0014[a]	0.00027	0.0033[a]	0.0015	0.0014[a]	0.00034	0.002[a]	0.00091[a]	0.025	0.018	0.029	0.013	0.021	0.003	0.005
Yb	0.0073	0.00072	0.022	0.0088	0.0092	0.0016	0.012	0.0071	0.15	0.12	0.18	0.085	0.13	0.016	0.005
Lu	0.0013[a]	0.00039	0.0029[a]	0.0012	0.0013[a]	0.00034	0.0021[a]	0.0012	0.022	0.017	0.025	0.012	0.018	0.0024	0.005
Y	0.1	0.015	0.42	0.19	0.11	0.032	0.11	0.025	2.8	0.95	4.2	0.98	3.3	0.69	0.005
ΣREY	0.95	0.11	4.9	2.4	1.08	0.28	1.02	0.1	45	12	72	22	64	15	
ΣLREE	0.78	0.089	4.2	2.1	0.89	0.23	0.76	0.087	41	12	65	21	58	14	
ΣHREE	0.17	0.025	0.66	0.3	0.19	0.049	0.26	0.047	4.6	1.6	6.6	1.5	5.3	1.1	

[a] Below LOQ, and replaced with LOQ.

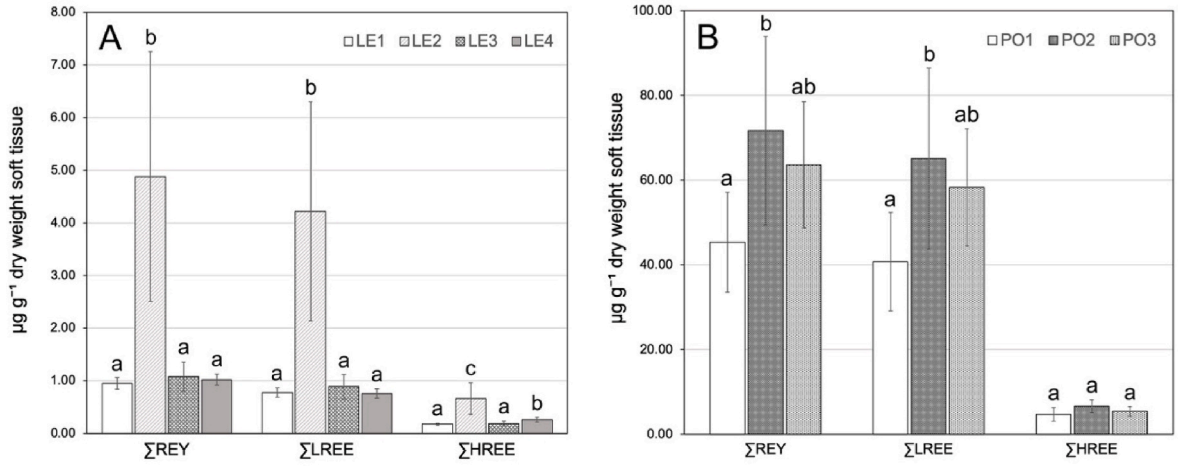


Fig. 2. Total (Σ) REY, LREE, and HREE concentrations (mean ± SD) in soft tissues of (A) *Mytilus* spp. from Lindesnes and (B) *Tapes* spp. from Porsgrunn in Southern Norway. LE1 and PO1 represent bivalves from the reference sites. Significant differences ($p < 0.05$) between sites are annotated by differing letters (Games-Howell *post-hoc* test).

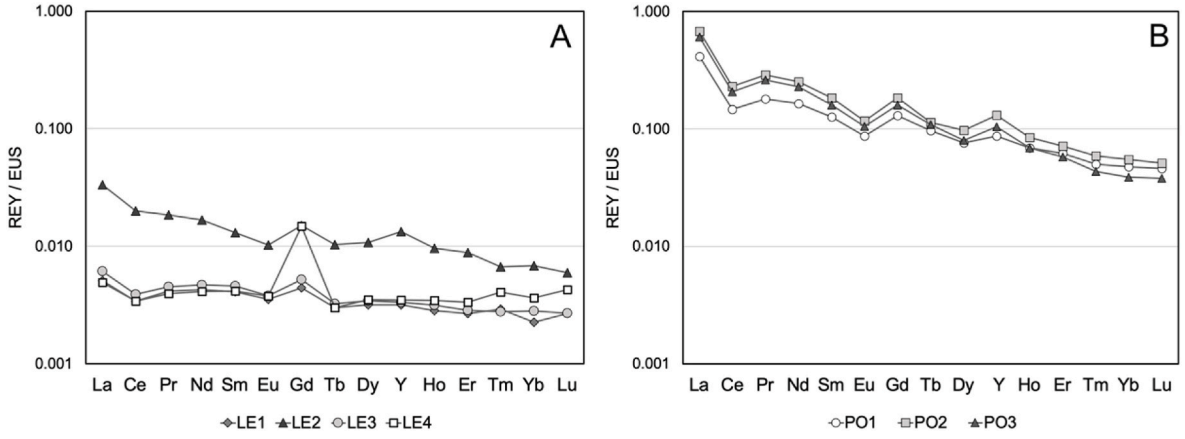


Fig. 3. Shale-normalized REY fractionation patterns in soft tissues of (A) *Mytilus* spp. from Lindesnes and (B) *Tapes* spp. from Porsgrunn in Southern Norway. LE1 and PO1 represent bivalves from the reference sites. Values are presented on a logarithmic scale.

4. Discussion

4.1. Total REY in the soft tissue of bivalves

Among the study sites in Lindesnes, only the Ramslandsvågen Bay site (LE2) showed a significantly elevated level of ΣREY in *Mytilus* spp. soft tissue compared to the background level. In comparison to other reports, mussel concentrations at all Lindesnes sites were still within the range of concentrations frequently reported for Mytilids worldwide, between 0.4 and 5 µg g^{-1} d.w. (see Supplementary S1) (MacMillan et al., 2017; Figueiredo et al., 2022; Briant et al., 2021; Costas-Rodríguez et al., 2010; Zhao et al., 2022). Nevertheless, ΣREY in Ramslandsvågen bay mussels (4.88 ± 2.37 µg g^{-1}) ranked amongst the higher range of concentrations so far reported in Mytilids, close to levels reported in mussels on the coast of the Guangdong province, China (4.96 µg g^{-1}) and in Quebec, Canada (5.16 µg g^{-1}) (Zhao et al., 2022; MacMillan et al., 2017). We attribute the high ΣREY concentration observed in this site (LE2) to the distinct characteristics of the substratum that the mussels were attached to. Compared to the other sites in Lindesnes, which had a rocky substrate, LE2 mussels were found in close association with sediment. LE2 mussels were thus potentially more exposed to sediment or suspended sediment particles which may carry sequestered REY.

Tapes spp. clams in Porsgrunn had the highest ΣREY concentrations (45–72 µg g^{-1}) so far reported in bivalve soft tissues to our knowledge (see literature comparison in Supplementary Fig. S2) (Akagi & Edanami, 2017; Wang et al., 2019; Ma et al., 2019; Briant et al., 2021; Budko et al., 2021; Barrat et al., 2022; Zhao et al., 2022). This displays the relatively high capacity of these clams for REY accumulation. However, this may also be related to possibly high concentrations of bioavailable REY in the area. The high ΣREY concentrations of the clams in Porsgrunn may be a result of strong freshwater influence in Frier fjord. The freshwater runoff in the fjord is completely dominated by the Skien River (with an annual mean discharge of 270 m^3 s^{-1}), which forms a brackish water layer at the fjord surface (Molvær, 1980). In rivers, much of the LREEs, which make up a large bulk of ΣREY, exist as organically stabilized colloids, and their presence in the dissolved REY pool can be affected by estuarine mixing (Sholkovitz, 1992). REY levels in rivers are known to decrease as they move from riverine to brackish to seawater, as a result of mixing processes with saltwater that cause the coagulation of colloidal and particulate-bound REY, leading to their subsequent sedimentation and high removal rates from water (Sholkovitz, 1992; Elderfield et al., 1990). With this, REY concentrations are expected to be greater in the brackish waters in Porsgrunn compared to the open sea in Lindesnes with little freshwater influence.

From a spatial perspective, in Porsgrunn there were higher ΣREY concentrations in the clams closer to the river mouth (PO2, PO3), followed by lower ΣREY (and ΣLREE) concentrations approaching the southern end of the fjord (PO1). As a highly industrialized catchment,

Frier Fjord has had historical pollution of nitrogen and phosphorous compounds, organic matter, metals and halogenated hydrocarbons due to effluents from pulp and chemical industries, and municipal wastewater (Molvær, 1980). In addition, erosion and the supply of polluted runoff from agricultural areas in Gjerpensdalen and Hovenga have also added significant quantities of nitrogen, phosphorus, and suspended material into the Skien River (Barland, 2005). While it is difficult to trace the contamination to a singular source of REY, the high concentrations at PO2 may derive from proximity to the waste and sources upstream of the Skien River or from the naturally higher REY concentrations from the river outflow.

The elevated concentrations in Porsgrunn compared to Lindesnes may also be related to the habitat variations between the two species of bivalves, which expose them to different environmental compartments. Mytilidae are epifauna, attached to solid exposed intertidal substrates such as rocks, while *Tapes* spp. on the other hand, as clams, are infauna, living on or buried in sand, mud, or gravel below the mid-tide level (Anacleto et al., 2016). While both these filter feeders have efficient sorting mechanisms for the particles they ingest (Jørgensen, 1981), clams that are directly exposed to contaminated sediment have been shown to significantly bioaccumulate REY in their soft tissue, as was the case for the Asian clam *Corbicula fluminea* (Bonnail et al., 2017). As sediment-burrowing bivalves, clams may also be exposed to pore waters, which contain higher REY concentrations than seawater and may exhibit different REY fractionation patterns (Haley et al., 2004).

Generally, LREE contributed to 82 ± 4% of the ΣREY concentration in *Mytilus* spp., and this proportion was found to be even greater in *Tapes* spp. (90 ± 3%). The elements La, Ce, and Nd were consistently the three most abundant REY, and accounted for approximately 75% and 84% of the average ΣREY in mussels and clams, respectively. Among the HREE, Y contributed the most, making up 9–11% and 5–6% of the ΣREY in mussels and clams, respectively.

Information on REY toxicity is still scarce, but one study has proven that La and Y exposure can be toxic to the embryonic development of mussels (*M. galloprovincialis*), with La having higher toxicity (EC50 ~ 50 μg L−1) than Y (EC50 800 μg L−1) (Mestre et al., 2019). In Andrade et al. (2023), environmentally relevant concentrations of Y (5 μg L−1) were able to induce lowered electron transport system activity, consumption of glycogen reserves, and activation of superoxide dismutase activity to avoid cellular damage in *M. galloprovincialis*. In *Ruditapes philippinarum*, Tb exposure has been shown to induce metabolic impairment and alterations in antioxidant capacity (Lompré et al., 2021) Future monitoring of seawater or sediment REY concentrations would allow for better evaluation of the exposure and risk of REYs to bivalves in Southern Norway.

4.2. Soft tissue REY fractionation patterns

The REY_{SN} composition in *Mytilus* spp. and *Tapes* spp. is characterized by LREE enrichment over HREE, with both having $[La/Yb]_{SN}$ ratios over 1. The relative LREE enrichment was higher in all *Tapes* spp., as seen from the steeper decline of the curve. LREE enrichment in soft tissue has previously been documented in clams (Wang et al., 2019; Zhang et al., 2009), mussels (MacMillan et al., 2017; Rodríguez-Velarte et al., 2022), and oysters (Ma et al., 2019; Zhao et al., 2022) around the world. REY_{SN} patterns of *Mytilus* spp. and *Tapes* spp. in this study resembled the soft tissue patterns of *Mytilus edulis* in arctic Canada (MacMillan et al., 2017) and *Ruditapes philippinarum* clams in Malauan Bay, China (Wang et al., 2019), which both featured negative Ce anomalies, positive Gd anomalies, and a constant depletion from LREE to HREE.

Considering the REY source to be the dissolved pool, Weltje et al. (2002) suggested that the higher solubility of LREE allows these to have higher bioavailability in biota. HREE on the other hand exhibit high lanthanide-ligand stability which, in the presence of available ligands from natural waters, could decrease HREE bioavailability and limit their bioaccumulation (Weltje et al., 2002). More recent studies however have started to emphasize the importance of sediment and suspended particles as important contributors to REY in bivalves, rather than the dissolved pool (Akagi and Edanami, 2017; Ma et al., 2019). This is evidenced by Tokyo Bay bivalves reflecting a similar REY pattern as their sediment (Akagi and Edanami, 2017), and the REY_{SN} of bivalves and sediment from the Chinese and Portuguese coasts all showing a common LREE enrichment (Zhao et al., 2022; Figueiredo et al., 2022). Barrat et al. (2022) suggested sedimentary organic matter as an important yet uninvestigated compartment, which may contain a substantial amount of readily-assimilable REY that can be absorbed with other nutrients by filter-feeding molluscs. With REY concentrations of 16–342 μg g^{-1} in estuarine and oceanic sedimentary organic matter, organic compounds can act as important REY scavenging phases in the aquatic environment (Freslon et al., 2014).

The negative Ce anomalies observed in Lindesnes and Porsgrunn bivalves may still point to a possible contribution of natural waters to the REY accumulation in soft tissue. Cerium is one of the two redox-sensitive LREE, and in the oxic conditions of the open ocean it is oxidized to the insoluble Ce(IV)-oxide phase which removes it from the solution, resulting in a Ce depletion that is a characteristic feature of seawater REY patterns (German & Elderfield, 1990). While the exact mechanisms of how REY anomalies occur in biota are still unclear, the poorer solubility of Ce^{4+} may also affect its bioavailability and result in its exclusion during assimilation by the filter-feeders.

4.3. Gd anomalies

In Lindesnes, a major finding of this study was the significantly large Gd anomaly ($Gd/Gd^* = 4.4$) observed in mussels southwest of the GBCA industry outfall (LE4), which was threefold higher than the rest of the sites. All bivalves in southern Norway, with the exception of mussels in LE4, only exhibited small, positive Gd anomalies in their soft tissue ($Gd/Gd^* = 1.24–1.41$; see Supplementary Table S4). The values of these Gd anomalies were either lower or comparable to those observed in the surrounding background seawater. Gd/Gd^* was 1.6 for open ocean water in the central North Atlantic, and 1.7–1.9 for coastal water in the southern North Sea (Ponnurangam et al., 2016; Kulaksız & Bau, 2007).

Several aspects make this large anomaly finding significant. Historically, monitoring of Gd in biota around the GBCA facility has been based on comparing the wet weight concentrations of Gd (μg g^{-1} w.w.) in biota collected around the discharge and a reference site. A monitoring program using *M. edulis* was already initiated in 2015 but was discontinued after finding similar Gd concentrations in mussels at all stations (DNV GL, 2016). In 2018, Gd was then measured from polychaetes from the soft-bottom sediment, but no clear concentration differences between stations could be deduced in part due to the limited sample material (DNV GL, 2019). Polychaetes were measured again in 2021 in more stations and Gd concentrations were found to have a 12 to 26-fold increase (DNV, 2022). This time however, Gd anomalies were calculated and the values reported ($Gd/Gd^* = 0.9–1.2$) were equivalent and even lower than the background, indicating no excess accumulation in soft-bottom fauna (DNV, 2022).

The use of Gd anomalies makes it possible to interpret anthropogenic Gd enrichment relative to the normal background concentrations, which are interpolated from the adjacent elements Sm and Tb (Ponnurangam et al., 2016). In our study, it allowed us to see that despite the higher Gd concentration (and total REY) in Ramslandvågen mussels (LE2), its Gd anomaly remained conserved and comparable to the background, indicating that Gd increased proportionally with higher total REY.

For LE4 mussels, a Gd anomaly of 4.4 indicates a source of excess Gd in the environment affecting the site. The GBCA production facility in Lindesnes releases pre-treated industrial spillwater containing traces of Gd in the form of gadodiamide at a depth of 40 m, and these emissions are estimated to affect an area of 500 m from the outlet into the water body (Miljødirektoratet, 2020). The water body of Mandal-Lindesnes

currently receives no other direct emissions from municipal sewage or the GBCA facility, and there is also no significant runoff from agriculture (DNV GL, 2019). In this area of southern Norway, the surface current flows from east to west towards Lindesnes Peninsula and continues along the coast (Norwegian Ministry of the Environment, 2013). With the westward flow of the current, the site most exposed to the emissions downstream of the spillwater pipe is LE4, southwest of the discharge. The facility reported periodic emissions with an expected maximum concentration of 0.005 tons of Gd-DOTA and gadodiamide per year for the period 2020–2030 (Miljødirektoratet, 2020).

The detection of this large anomaly also indicates mussels may be more suitable indicators compared to polychaetes which were sampled in the same year. No data on Gd in biota has been reported southwest of the discharge in the previous 2015, 2018, and 2021 monitoring programs, which might also explain the lack of findings in these assessments. Overall, the results serve as a recommendation to include this SW site and reimplement mussel monitoring in Lindesnes.

5. Conclusion

To our knowledge, this study represents the first assessment of REY in bivalves in Norway. It demonstrated the use of *Mytilus* spp. and *Tapes* spp. as sentinels of REY contamination originating from different anthropogenic activities along the southern coast of Norway.

Overall, the shale-normalized REY patterns in both bivalves were characterized by LREE enrichment over HREE, a negative Ce anomaly, and a positive Gd anomaly. The LREE enrichment suggests that the sediment and suspended particles may be important contributors to REY in bivalves, rather than the dissolved pool. In Lindesnes, *Mytilus* spp. downstream of the spillwater pipe had shown a footprint of GBCA production in the form of a significantly large, positive Gd anomaly that indicated excess bioavailable Gd in the area, which has not been reported in previous monitoring programs. This study presents a strong case to reimplement biomonitoring in Lindesnes using mussels as a more suitable indicator, and to monitor the area SW of the discharge. In the region of Porsgrunn, *Tapes* spp. Bioaccumulated very high levels of REY in their soft tissue, with clams closer to the river mouth having significantly elevated concentrations over the background. The overall higher REY concentrations in clams compared to mussels possibly reflected the effect of habitat variations between the bivalves, and the strong riverine influence on Frier fjord, which may cause greater bioavailability of dissolved REY.

For future research, it is also recommended to conduct a spatial analysis of the REY concentrations in the different environmental compartments in these study areas, with a special focus on sediment, and suspended particulate organic and inorganic matter. This would contribute to a better understanding of the critical factors that give rise to the REY fractionation patterns observed in bivalves for future biomonitoring studies.

Credit roles

Lyen Castro: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Validation, Visualization, Writing – original draft, Writing – review & editing. **Julia Farkas:** Conceptualization, Formal analysis, Funding acquisition, Project administration, Validation, Resources, Writing – review & editing. **Bjørn Munro Jenssen:** Conceptualization, Formal analysis, Resources, Supervision, Writing – review & editing. **Stefania Piarulli:** Methodology, Writing review & editing. **Tomasz Ciesielski:** Conceptualization, Formal analysis, Investigation, Resources, Supervision, Validation, Writing – review & editing.

Funding sources

This work was supported by the Norwegian Research Council [Grant Agreement No. 301236] under Project ELEMENTARY.

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

Data will be made available on request.

Acknowledgements

This is to acknowledge Achillefs Zervos for field sample collection of the bivalves, Dr. Anica Simic, Dr. Kyyas Seyitmuhammedov, and Sara Cvetkovics for technical assistance in sample preparation and processing and Marianne Steinsvik Kjos for ICP-MS determination.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2023.122300.

References

Akagi, T., Edanami, K., 2017. Sources of rare earth elements in shells and soft-tissues of bivalves from Tokyo Bay. Mar. Chem. 194, 55–62. https://doi.org/10.1016/j.marchem.2017.02.009.

Anacleto, P., Maulvault, A.L., Barbosa, V., Nunes, M.L., Marques, A., 2016. Shellfish: Characteristics of Crustaceans and Mollusks.

Barrat, J.A., Chauvaud, L., Olivier, F., Poitevin, P., Bayon, G., Salem, D.B., 2022. Rare earth elements and yttrium in suspension-feeding bivalves (dog cockle, *Glycymeris glycymeris* L.): accumulation, vital effects and pollution. Geochem. Cosmochim. Acta. https://doi.org/10.1016/j.gca.2022.10.033.

Barland, K., 2005. Regional Overvåkning Av Vannforekomster I Telemark 1970-2005.

Bau, M., Dulski, P., 1996. Anthropogenic origin of positive gadolinium anomalies in river waters. Earth Planet Sci. Lett. 143 (1–4), 245–255. https://doi.org/10.1016/0012-821X(96)00127-6.

Bau, M., Schmidt, K., Pack, A., Bendel, V., Kraemer, D., 2018. The European Shale: an improved data set for normalisation of rare earth element and yttrium concentrations in environmental and biological samples from Europe. Appl. Geochem. 90, 142–149. https://doi.org/10.1016/j.apgeochem.2018.01.008.

Beyer, J., Green, N.W., Brooks, S., Allan, I.J., Ruus, A., Gomes, T., Bråte, I.L.N., Schøyen, M., 2017. Blue mussels (*Mytilus edulis spp.*) as sentinel organisms in coastal pollution monitoring: a review. Mar. Environ. Res. 130, 338–365. https://doi.org/10.1016/j.marenvres.2017.07.024.

Bonnail, E., Pérez-López, R., Sarmiento, A.M., Nieto, J.M., DelValls, T.Á., 2017. A novel approach for acid mine drainage pollution biomonitoring using rare earth elements bioaccumulated in the freshwater clam Corbicula fluminea. J. Hazard Mater. 338, 466–471.

Briant, N., Le Monier, P., Bruzac, S., Sireau, T., Araújo, D.F., Grouhel, A., 2021. Rare earth element in bivalves' soft tissues of French metropolitan coasts: spatial and temporal distribution. Arch. Environ. Contam. Toxicol. 81 (4), 600–611. https://doi.org/10.1007/s00244-021-00821-7.

Budko, D.F., Lobus, N.V., Vedenin, A.A., 2021. Dataset on the content of major, trace, and rare-earth elements in the bottom sediments and bivalve mollusks of the Kara Sea (Arctic Ocean). Data Brief 36, 107087.

Castor, S.B., Hedrick, J.B., 2006. Industrial Minerals & Rocks: Commodities, Markets, and Uses, vol. 7. Society for Mining, Metallurgy, and Exploration.

Coryell, C.D., Chase, J.W., Winchester, J.W., 1963. A procedure for geochemical interpretation of terrestrial rare-earth abundance patterns. J. Geophys. Res. 68 (2), 559–566. https://doi.org/10.1029/JZ068i002p00559.

Costas-Rodríguez, M., Lavilla, I., Bendicho, C., 2010. Classification of cultivated mussels from Galicia (Northwest Spain) with European Protected Designation of Origin using trace element fingerprint and chemometric analysis. Anal. Chim. Acta 664 (2), 121–128. https://doi.org/10.1016/j.aca.2010.03.003.

DNV GL, 2016. Miljøovervåking Ramslandsvågen Og Utenforliggende Områder. Report 2015. Report number: 2016- 0179, Rev 1.

DNV GL, 2019. Miljøovervåking Utenfor Ramslandsvågen 2018. Report Number: 2019-0060, Rev 0.

DNV, 2022. Resipientundersøkelse Utenfor Ramslandsvågen 2021, GE Healthcare AS, Lindesnes Factories. Report number: 2021-0881, Rev 1.

Elderfield, H., Upstill-Goddard, R., Sholkovitz, E.R., 1990. The rare earth elements in rivers, estuaries, and coastal seas and their significance to the composition of ocean waters. Geochem. Cosmochim. Acta 54 (4), 971–991.

Figueiredo, C., Oliveira, R., Lopes, C., Brito, P., Caetano, M., Raimundo, J., 2022. Rare earth elements biomonitoring using the mussel *Mytilus galloprovincialis* in the Portuguese coast: seasonal variations. Mar. Pollut. Bull. 175, 113335 https://doi.org/10.1016/j.marpolbul.2022.113335.

Freslon, N., Bayon, G., Toucanne, S., Bermell, S., Bollinger, C., Chéron, S., et al., 2014. Rare earth elements and neodymium isotopes in sedimentary organic matter. Geochem. Cosmochim. Acta 140, 177–198.

German, C.R., Elderfield, H., 1990. Application of the Ce anomaly as a paleoredox indicator: the ground rules. Paleoceanography 5 (5), 823–833.

Gonzalez, V., Vignati, D.A.L., Leyval, C., Giamberini, L., 2014. Environmental fate and ecotoxicity of lanthanides: are they a uniform group beyond chemistry? Environ. Int. 71, 148–157. https://doi.org/10.1016/j.envint.2014.06.019.

Gwenzi, W., Mangori, L., Danha, C., Chaukura, N., Dunjana, N., Sanganyado, E., 2018. Sources, behaviour, and environmental and human health risks of high-technology rare earth elements as emerging contaminants. Sci. Total Environ. 636, 299–313. https://doi.org/10.1016/j.scitotenv.2018.04.235.

Haley, B.A., Klinkhammer, G.P., McManus, J., 2004. Rare earth elements in pore waters of marine sediments. Geochem. Cosmochim. Acta 68 (6), 1265–1279.

Haxel, G., 2002. Rare earth elements: critical resources for high technology. US Department of the Interior, US Geological Survey 87 (No. 2).

IBM Corp, 2021. IBM SPSS Statistics for Macintosh. Released. IBM Corp, Armonk, NY. Version 29.

Jørgensen, C., 1981. Feeding and cleaning mechanisms in the suspension feeding bivalve Mytilus edulis. Mar. Biol. 65 (2), 159–163.

Kulaksız, S., Bau, M., 2007. Contrasting behaviour of anthropogenic gadolinium and natural rare earth elements in estuaries and the gadolinium input into the North Sea. Earth Planet Sci. Lett. 260 (1–2), 361–371. https://doi.org/10.1016/j.epsl.2007.06.016.

Le Goff, S., Barrat, J.-A., Chauvaud, L., Paulet, Y.-M., Gueguen, B., Ben Salem, D., 2019. Compound-specific recording of gadolinium pollution in coastal waters by great scallops. Sci. Rep. 9 (1), 8015. https://doi.org/10.1038/s41598-019-44539-y.

Lerat-Hardy, A., Coynel, A., Dutruch, L., Pereto, C., Bossy, C., Gil-Diaz, T., Capdeville, M.-J., Blanc, G., Schäfer, J., 2019. Rare Earth Element fluxes over 15 years into a major European Estuary (Garonne-Gironde, SW France): hospital effluents as a source of increasing gadolinium anomalies. Sci. Total Environ. 656, 409–420. https://doi.org/10.1016/j.scitotenv.2018.11.343.

Lompré, J.S., Moleiro, P., De Marchi, L., Soares, A.M., Pretti, C., Chielini, F., et al., 2021. Bioaccumulation and ecotoxicological responses of clams exposed to terbium and carbon nanotubes: comparison between native (Ruditapes decussatus) and invasive (Ruditapes philippinarum) species. Sci. Total Environ. 784, 146914.

Ma, L., Dang, D.H., Wang, W., Evans, R.D., Wang, W.X., 2019. Rare earth elements in the Pearl River Delta of China: potential impacts of the REE industry on water, suspended particles and oysters. Environ. Pollut. 244, 190–201.

MacMillan, G.A., Chételat, J., Heath, J.P., Mickpegak, R., Amyot, M., 2017. Rare earth elements in freshwater, marine, and terrestrial ecosystems in the eastern Canadian Arctic. Environ. Sci.: Process. Impacts 19 (10), 1336–1345. https://doi.org/10.1039/C7EM00082K.

Mestre, N.C., Sousa, V.S., Rocha, T.L., Bebianno, M.J., 2019. Ecotoxicity of rare earths in the marine mussel *Mytilus galloprovincialis* and a preliminary approach to assess environmental risk. Ecotoxicology 28 (3), 294–301.

Migaszewski, Z.M., Gałuszka, A., 2015. The characteristics, occurrence, and geochemical behavior of rare earth elements in the environment: a review. Crit. Rev. Environ. Sci. Technol. 45 (5), 429–471. https://doi.org/10.1080/10643389.2013.866622.

Miljødirektoratet, 2020. Søknad Om Økt Produksjon Ved GE Healthcare AS. *Lindesnes* (No. 2019/299). Miljødirektoratet. https://www.miljodirektoratet.no/hoeringer/2020/april-2020/ge-healthcare-lindesnes-soker-om-endret-tillatelse-/.

Molvær, J., 1980. Deep water renewals in the Frierfjord-an intermittently anoxic basin. In: Fjord Oceanography. Springer, Boston, MA, pp. 531–537.

Norwegian Ministry of the Environment, 2013. Integrated Management of the Marine Environment of the North Sea and Skagerrak (Management Plan).

Nuss, P., Blengini, G.A., 2018. Towards better monitoring of technology critical elements in Europe: coupling of natural and anthropogenic cycles. Sci. Total Environ. 613–614, 569–578.

Pereto, C., Coynel, A., Lerat-Hardy, A., Gourves, P.Y., Schäfer, J., Baudrimont, M., 2020. Corbicula fluminea: a sentinel species for urban Rare Earth Element origin. Sci. Total Environ. 732, 138552.

Piarulli, S., Hansen, B.H., Ciesielski, T., Zocher, A.-L., Malzahn, A., Olsvik, P.A., Sonne, C., Nordtug, T., Jenssen, B.M., Booth, A.M., Farkas, J., 2021. Sources, distribution and effects of rare earth elements in the marine environment: current knowledge and research gaps. Environ. Pollut. 291, 118230 https://doi.org/10.1016/j.envpol.2021.118230.

Ponnurangam, A., Bau, M., Brenner, M., Koschinsky, A., 2016. Mussel shells of *Mytilus edulis* as bioarchives of the distribution of rare earth elements and yttrium in seawater and the potential impact of pH and temperature on their partitioning behavior. Biogeosciences 13 (3), 751–760. https://doi.org/10.5194/bg-13-751-2016.

Rodríguez-Velarte, P., Babarro, J.M.F., Cobelo-García, A., 2022. Bioaccumulation patterns of trace elements by native (M. galloprovincialis) and invasive (X. securis) mussels in coastal systems (Vigo Ria, NW Iberian Peninsula). Mar. Pollut. Bull. 176, 113463 https://doi.org/10.1016/j.marpolbul.2022.113463.

Staalstrøm, A., 2013. *Overvåkning I Forbindelse Med Rehabilitering Av Knardalstrand Renseanlegg (*NIVA Rapport 6529). Norwegian Institute for Water Research. http://hdl.handle.net/11250/216367.

Sholkovitz, E.R., 1992. Chemical evolution of rare earth elements: fractionation between colloidal and solution phases of filtered river water. Earth Planet Sci. Lett. 114 (1), 77–84.

Trapasso, G., Chiesa, S., Freitas, R., Pereira, E., 2021. What do we know about the ecotoxicological implications of the rare earth element gadolinium in aquatic ecosystems? Sci. Total Environ. 781, 146273 https://doi.org/10.1016/j.scitotenv.2021.146273.

Wang, Z., Yin, L., Xiang, H., Qin, X., Wang, S., 2019. Accumulation patterns and species-specific characteristics of yttrium and rare earth elements (YREEs) in biological matrices from Maluan Bay, China: implications for biomonitoring. Environ. Res. 179, 108804 https://doi.org/10.1016/j.envres.2019.108804.

Zepf, V., 2013. Rare earth elements: what and where they are. In: Zepf, V. (Ed.), Rare Earth Elements. Springer Berlin Heidelberg, pp. 11–39. https://doi.org/10.1007/978-3-642-35458-8_2.

Zhang, H., Deng, L., Yang, J., Zhiguo, S., Jianbin, X., 2009. Rare earth elements in marine organisms from Shenzhen coastal region. Wei Sheng Yan Jiu = J. Hyg. Res. 38 (5), 543–545.

Zhao, P., Bi, R., Sanganyado, E., Zeng, X., Li, W., Lyu, Z., et al., 2022. Rare earth elements in oysters and mussels collected from the Chinese coast: bioaccumulation and human health risks. Mar. Pollut. Bull. 184, 114127 https://doi.org/10.1016/j.marpolbul.2022.114127.