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Biogeochemical impact of historical submarine mine tailings on benthic ecosystems in the Repparfjord (Northern Norway)

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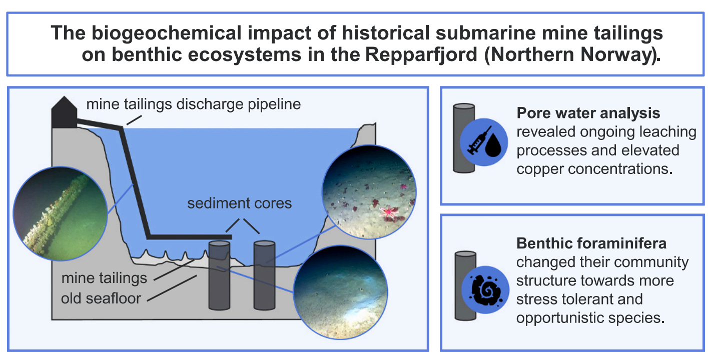
HIGHLIGHTS

- Benthic copper release from submarine mine tailing disposal site is ongoing.
- Natural bedrock contributes to high Ni, Pb, and Zn pore water values.
- Stress tolerant and opportunistic foraminifera in post-tailing sediment.
- Paleo-environmental study crossing the tailing disposal period using foraminifera.

GRAPHICAL ABSTRACT



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ABSTRACT

Historical copper mine tailings deposited in the Repparfjord, Northern Norway, provided new insight into the biogeochemical impact of submarine tailings disposals on high-latitude coastal ecosystems. The submarine tailings disposal in the Repparfjord represents a product of mining activities between 1972 and 1979. Their environmental impact has been extensively studied during the last decade, but geochemistry of the sediment pore water, which is crucial to assess and monitor the in-situ metal leaching and bioavailability, has never been analysed. The actual impact on the benthic fauna remains poorly known. Therefore, this study couples the pore water chemistry and the foraminiferal analysis obtained from selected sediment cores (gravity core, multicore, box cores) to examine metal stability and the past and current status of the foraminifera community. We measured down-core sulfate and trace metal concentrations and Eh-Ph and applied the Shannon index, the AZTI's Marine Biotic Index (F-AMBI) index and the foraminiferal abnormality index. This study confirms the ongoing leaching of Cu from the underlying mine tailings and release across the sediment-water interface. Leaching of Ni, Zn and Pb have been attributed to weathering of natural bedrock lithologies. The original benthic foraminiferal community disappeared almost entirely during the disposal period, and now it is dominated by stress-tolerant

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and opportunistic species like *Bulimina marginata* and *Spiroplectammina biformis*. Anyhow, against previous assumptions, the community composition changed, while the overall diversity and abnormalities (FAI) shell formation is unaffected by elevated Cu concentrations.

1. Introduction

The mining industry is considered the world's largest waste producer, and providing safe disposal sites for mining waste poses significant environmental challenges. While the submarine disposal of mining wastes is prohibited in many places, it is still practised in some countries (Dold, 2014; Ramirez-Llodra et al., 2015). Submarine tailings disposal (STD) was often considered a convenient way to dispose of unwanted mining material in countries with predominantly mountainous landscapes unfavourable for safe disposal on land, such as Norway. It has, however, become substantial to provide safe storage with minimised risks for society and the environment. Leaching of trace metals into the surrounding environment has repeatedly been reported at former STD sites (Koski, 2012; Ramirez-Llodra et al., 2015; Schaanning et al., 2019). Sulfide minerals, that used to be stable within the ore, are at risk of oxidation and alteration into more soluble forms once they are deposited in a marine environment. Even though the availability of oxygen within the sediment is limited, processes like vertical mixing of tailings affected sediment layers driven by bioturbation or upward diffusion within the sediment column can expose those minerals to oxidising conditions (Amato et al., 2016; McDonald et al., 2010). Measuring depth profiles of dissolved trace metals, pH and redox potential within the sediment pore waters has been suggested to be suitable for monitoring biogeochemical processes in the sediment of mine tailing-affected areas (McDonald et al., 2010). Still, the potentially severe environmental impact of STDs is often at risk of being underestimated, as in many of the biogeochemical processes within the sediments, the interactions and effects of tailings material on the seafloor ecosystems are not fully understood. Benthic foraminifera (Rhizaria) are part of the meiofauna and can be used to assess biological impact. Due to their high sensitivity to environmental change, they have become good bioindicators of natural stress and human disturbance, particularly in coastal areas such as fjords (Alve et al., 2016; Klootwijk et al., 2021). Moreover, their high preservation potential makes it possible to track changes in assemblage composition and diversity that may reflect the influence of past anthropogenic activities (Frontalini and Coccioni, 2008; Dijkstra et al., 2017).

Historical STDs in Norway provide an excellent natural laboratory for investigating the long-term stability of tailing materials in submarine environments and their impact on high-latitude benthic ecosystems. Previous studies in Repparfjord have shown limited lateral dispersion of tailing particles due to the fjord morphology (Pedersen et al., 2018; Sternal et al., 2017), but also indicated high amounts of Cu, Cr, and Ni exceeding the national sediment contamination thresholds (Mun et al., 2020b). Mineralogical analyses on the historical tailings and thermodynamic modeling suggested that Cu is predominantly hosted in crystal lattices of sulfide minerals (Mun et al., 2021). Permanent exposure of the benthic fauna in the Repparfjord to elevated Cu concentrations was proposed by Sternal et al. (2017), and the current status of the benthic macrofauna have been recently investigated by Trannum et al. (2023). Pore water data from Repparfjord are currently lacking, thus preventing any quantitative assessment of STD stability. To fill those gaps, we conducted a multidisciplinary research expedition in 2022, during which we collected sediment samples for pore water analyses and foraminiferal analysis and acquired videos of the seafloor with a towed camera system for the first time in a Norwegian mine-impacted fjord. The goals of this study are i) to quantify metal fluxes in the sediment using downcore pore water concentration profiles, ii) to assess the present-day and past diversity and composition of the benthic foraminiferal community by applying diverse foraminiferal-based indexes, iii) visually explore the status of the seafloor ecosystems using seafloor imagery. The obtained results should contribute to future tailing management plans and improve mitigation of environmental challenges in the Arctic.

2. Study area

Repparfjord, a fjord located in the Hammerfest municipality, northern Norway, stretches approximately 13 km in length and 4 km in width (Fig. 1). It is relatively shallow, with an average depth of 50 m and a maximum depth of 123 m. The fjord consists of a deeper outer basin and a shallower inner basin, separated by a northeast-southwest oriented sill located at a water depth of 50 m (Fig. 1; Andersson et al., 2018; Sternal et al., 2017). Due to the sill and the narrow morphology of the fjord, the exchange of water and sediment between the outer and inner basins is limited, especially below sill depth. Natural sediments in the Repparfjord are characterized by a significant contribution of fluvial material brought by the river of Repparfjordelva (e.g., Sternal et al., 2017). The river predominantly drains Paleoproterozoic basement rocks (mafic volcanics intercalated with sedimentary carbonate-siliciclastic rocks) of the Repparfjord Tectonic Window (RTW). The RTW hosts numerous Cu occurrences, including the Nussir and Ulveryggen sediment-hosted Cu deposits (Mun et al., 2020a; Torgersen et al., 2016). The Ulveryggen deposit was extensively mined between 1972 and 1979 and approximately 3 million tons of Cu ore were extracted (Sternal et al., 2017), followed by the discharge of 1 million tons of mine tailings at the inner fjord (Kvassnes and Iversen, 2013). Future mining activities at the historical Ulveryggen mine and a new site at the Nussir deposit (Nussir ASA, Norway) require a new disposal site and therefore a confined outer area of the Repparfjord has been allocated for storage of 30 million tons of mine tailings (Fig. 1; Christensen et al., 2011).

3. Material and methods

The sampling campaign was conducted during the CAGE22-1 expedition in early April 2022 using the UiT's research vessel RV Helmer Hanssen (Argentino et al., 2022). Sediment sampling sites (Fig. 1, Table 1) were selected based on a video survey of the seafloor with a DSPL Multi SeaCam HD camera and their locations related to the distribution of historic STD. At the same time, seabed mapping was conducted with the Kongsberg Simrad EM302 multibeam sonar system (Argentino et al., 2022).

3.1. Sampling of marine sediments

Different methods of sediment coring were applied, depending on the properties of the seafloor. Cores 1 and 2 (Fig. 1) were obtained with a KC Denmark DK8000 multicorer. Because a hard crust in the tailings affected area hindered the multicorer from penetrating, a box corer was used as an alternative for cores 4, 5, 6 and 7. It consists of a 50 × 50 × 50 cm steel sampling box from which the sediment cores were subsampled manually by pushing plastic liners, identical to the ones used for the multicorer, into the sediment immediately after the recovery. All plastic liners were predrilled with a vertical line of small holes in 1 or 2 cm steps for later pore water extraction and sealed with duct tape. Additionally, a gravity corer with a 1.3 ton head weight was used for core 3. This core was used for porosity determination (tailing interval and natural sediment) and for the analysis of benthic foraminifera from above and below the mine tailings layer.

3.2. Pore water and gas sampling, analysis and data processing

Pore water investigations were conducted on cores 1 (historic tailing area), 4 (river delta) and 6 (historic tailing area). Pore water samples were collected with 0.15 μm Rhizon filters through the predrilled holes in the plastic liners. Each sample was split into two aliquots: (1) for the analysis of trace metals, 3 mL of the obtained sample were transferred to Eppendorf tubes and acidified with 10 μL Suprapur 60 % HNO_3 onboard. Onshore, 500 μL of each sample were diluted with 9.2 mL of Milli-Q water, 200 μL HNO_3 and 100 μL indium solution as internal standard. The concentration of trace metals (Al, As, Ba, Cd, Co, Cr, Cu, Fe, Li, Mn, Mo, Ni, Pb, Rb, Sb, Se, Sn, Sr, Ti, U, V, Zn) was determined with a Thermo Element 2 HR-ICP-MS at the Ruđer Bošković Institute in Zagreb, Croatia. GEMS/Water intercalibration standards were run throughout the session and yielded absolute differences from the certified values better than 0.31 μg/L (ppb). Blanks were better than 0.09 μg/L (ppb) for all the elements. (2) For the analysis of sulfate (SO_4^{2-}), the remaining pore water samples were kept in Eppendorf tubes and stored at −20 °C until the SO_4^{2-} concentration was measured via ion chromatography on a Metrosep A Supp 4 column at University of Bergen (Norway). Repeated measurements of intercalibration ERM® CA016a certified material were used to check accuracy and precision during the analyses; measured values agree within the certified uncertainty (±10 mg/L; 0.9 mM). Aliquots of pore waters were used for onboard pH-Eh analysis with a HACH HQ440D multimeter. Bottom water was collected directly from the multicore using a Rhizon filter. Cores 2, 3 and 5 were additionally sampled for headspace gas by transferring bulk sediment samples into a 20 mL serum vial containing 5 mL of 1 M NaOH. Those samples were measured with a ThermoScientific GC Trace 1310 gas chromatograph-mass spectrometer equipped with a Restek Rt-Alumina BOND/Na_2SO_4 column for hydrocarbon gas separation hosted at Geolab, Department of Geosciences in Tromsø (Norway).

Diffusive fluxes of metals across the sediment-water interface as well as consumption and production rates in the sediment were quantified by applying a one-dimensional diagenetic transport-reaction model on the obtained pore water metal profiles (Boudreau, 1997). The model was implemented on the PROFILE software (version 1.0) (Berg et al., 1998). It assumed steady state conditions and was corrected for tortuosity according to Iversen and Jørgensen (1993). The input porosity was determined from weight loss upon drying of a known volume of sediment (5 mL). Porosity was 0.47 for the mine tailings and 0.51 for the natural sediment. Diffusion coefficients of metals and sulfate were extracted from the "Marelac" package for R (Soetaert and Petzoldt, 2020) for in-situ bottom water temperature of 2.2 °C and salinity of 34.1 psu recorded by CTD measurements. In addition to the fluxes at the sediment-water interface determined by the model, linear fluxes were calculated for comparison, assuming linear gradient in the upper zone and applying Fick's first law of diffusion and Boudreau's correction for tortuosity in sediments (Schulz, 2006). There is currently no environmental classification available for pore waters so we tentatively assessed

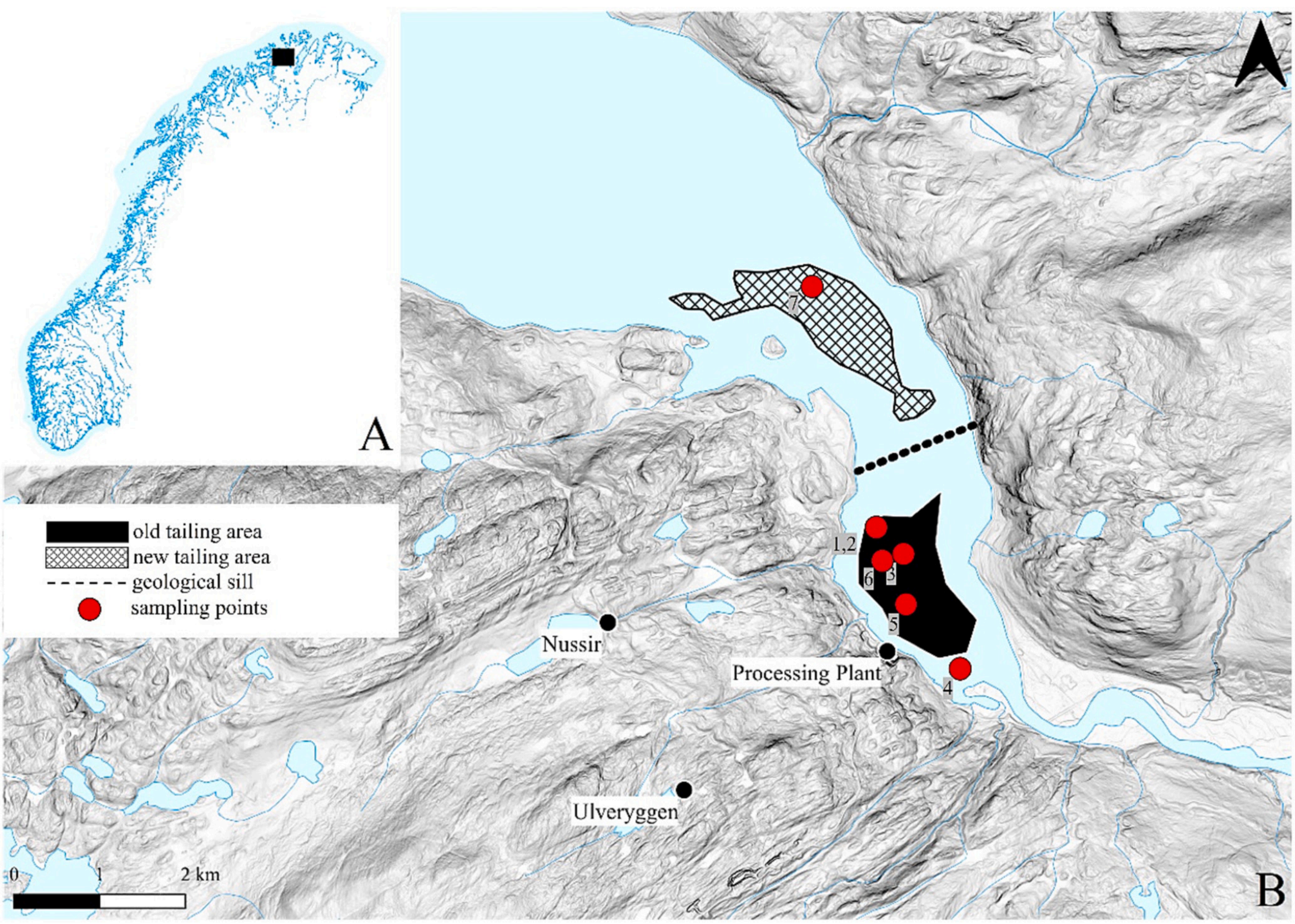

Fig. 1. Overview map of Norway (A) The research area is indicated by a black square. Regional map of the Repparfjord (B) including the location of the geological sill (black dashed line), the historical mine tailings (black polygon, "old tailing" in the map) modified from Mun et al. (2020b) and the planned disposal area ("new tailing area" in the map) for the new tailings (checkered polygon), the locations of the Nussir and Ulveryggen ores and the former processing plant according to Nussir ASA. Sampling points in the mine tailings disposal area (1,2,3,5 and 6), in the reference area in the inner fjord (4) and in the outer fjord (7). Topographic map from kartverket.no.

the water quality using the Norwegian environmental quality classification of metal contaminants in coastal waters defined by the Norwegian pollution control authority Miljødirektoratet (Guideline M-608, 2016). As discussed later in the text, this classification might overestimate the degree of contamination.

3.3. Benthic foraminifera sampling, sample analysis and data processing

The first 5 cm of the cores 2, 5 and 7 were sampled onboard in cm-steps for the analysis of benthic foraminifera. While we can assume the pore water conditions measured in cores 1 and 6 (tailing area) to be representative of cores 2 (same box core as core 1; Table 1) and 5, we do not have pore water datasets from the outer fjord where core 7 was collected. To preserve and stain the living organisms, the samples were stored in a 2 gL^{-1} solution of rose Bengal in 96 % ethanol for at least 14 days. Additionally, the gravity core 3 was sampled in 1 cm steps at the top (1–4 cm) and 5 cm steps throughout the complete retrieved length to assess the sedimentary archive. To compare the past and current community structure, the total assemblages of benthic foraminifera (i.e. living and dead ones) were considered. For each sample, up to 300 specimens of the 125–500 μm fraction were picked and identified to species level or lowest taxon possible according to Schönfeld et al. (2012), while the percentage of foraminifera with abnormal tests (Coccioni and Frontalini, 2005) was noted as well. Several samples contained <300 specimens. Here, all the specimens present in one sample were counted. The nomenclature of foraminiferal species follows the accepted species names published in the World Register of Marine Species (WoRMS) database (WoRMS Editorial Board, 2023).

The $EcoQS_{bf(H)}$ of the benthic foraminiferal communities was determined based on the guidelines of the European Water Framework Directive (WFD) (European Parliament, 2000) and following the classification scheme of Bouchet et al. (2012) (Table 2). They recommend applying the bias corrected version of the Shannon index H'bc (Chao and Shen, 2003) on the absolute species numbers of each sample, which has been confirmed to be suitable for reconstructing the ecological status by several studies (Dijkstra et al., 2017; Dolven et al., 2013). All calculations were performed using the “Entropy” library version 1.3.1 in R (Hausser and Strimmer, 2008). To not only compare the diversity of the samples for benthic foraminifera as a univariate descriptor, but also the composition of the communities, non-metric multidimensional scaling (NMDS) was applied, using the “vegan” package in R (Oksanen et al., 2022). Hereby, the information from multiple dimensions (sampling site, sampling depth and number of counted specimens per species in each sample) was collapsed into two dimensions using rank orders to allow for visualization and interpretation. An $R^2 > 0.9$ of both, the non-metric fit and the linear fit of the regression between the interpoint distances ensured that original dissimilarities were preserved during the reduction of dimensions. Additionally, the F-AMBI index, an internationally established marine biotic index adapted for benthic foraminifera, was determined based on the grouping scheme of Alve et al. (2016) and Bouchet et al. (2021). They assigned different species of foraminifera in Norwegian fjords to five groups along a total organic carbon (TOC) gradient, each representing the sensitivity of the resembling species to organic matter enrichment. The F-AMBI values were also assigned to different $EcoQS_{bf(F\text{-}AMBI)}$ classes based on the classification scheme of Bouchet et al. (2021) (Table 2). This allowed a meaningful interpretation of the different assemblage clusters revealed in the NMDS analysis. The ecological quality ratio (EQR) was calculated to evaluate EcoQS based on in situ reference condition. EQR represents the relationship between a biological parameter, such as diversity (Exp (H)), and the values observed under the reference condition. The WFD defines as a reference condition the biological quality at a high status (European Parliament). The EQR ranges from 0 (poor EcoQS) to 1 (high EcoQS) and the class boundary values are shown in Table 2.

Table 2
Criteria for determining the EcoQSrequired by the WFD classification and the corresponding MSFD classes “good” and “bad” using 125–500 μm dry picked foraminifera (Bouchet et al., 2012; Bouchet et al., 2021). The ecosystem impact was additionally defined by Dijkstra et al. (2017). Class boundaries values for Exp(H′), F-AMBI and EQR.

EcoQS	Ecosystem impact	Exp(H) ($EcoQS_{bf(H)}$)	F-AMBI ($EcoQS_{bf(F\text{-}AMBI)}$)	EQR
Bad	Severe acute impact	< 2.5	5.5–7	0–0.2
Poor	Acute impact	2.5–5	4.3–5.5	0.2–0.4
Moderate	Chronic impact	5–7.5	3.3–4.3	0.4–0.6
Good	No impact	7.5–10	1.2–3.3	0.6–0.8
High	Reference conditions	> 10	0–1.2	0.8–1

4. Results and discussion

4.1. Seafloor observations and sediment lithology

The towed camera videos acquired across the historical tailing disposal site revealed the presence of tailing material exposed at the seafloor (Fig. 2A, B). The tailing material appears as light grey/brown in color, forming patches up to a few meters across and up to half a meter tall. We observed bioturbation traces on the top, but no macrofauna was clearly seen on the tailing. On the surrounding dark brown sandy-mud sediments, large areas covered by tubeworms are observed, with sporadic sea stars and anemones colonizing larger pebbles. The tubeworms stick out of the seafloor for up to 4–5 cm and onboard observations indicated that they are ca 10 cm in length and have a diameter of ca. 2 mm (Argentino et al., 2022). During the dive, we found an old pipeline (Fig. 2 C), now densely colonized by sessile organisms. The precise classification of the macrofauna is out of the scope of this study and was discussed in Trannum et al. (2023). The videos that we acquired will be analysed by future research. The sill area is characterized by a different sediment grain size at the seafloor, including gravel and pebbles colonized by red-pink algae and other sessile organisms, mostly anemones (Fig. 2 D). Other organisms observed during the video surveys are sponges, brittle stars, bivalves, gastropods, fishes, and sea urchins. The outer fjord is characterized by a muddy seafloor with numerous fish holes and some biogenic trails (Fig. 2E, F). Fishes are more abundant and diverse compared to other areas, with the presence of flat fishes and red fishes. Anemones are rare, and fewer pebbles were observed.

Table 1
List of sediment cores used either for pore-water (PW) analysis or foraminiferal assemblages (F). Core numbers will be used in this study, while the original core numbers are unique to every sample taken with RV Helmer Hanssen. Abbreviations in the original core no. refer to multicore (MC), gravity core (GC) and box core (BC), while A, B and C stand either for one of the cores obtained within one multi core dive or for one of the cores subsampled from the box corer.

Core no.	Original core no.	Recovery [cm]	Water depth [m]	PW	F	Latitude	Longitude
1	CAGE-22-1-HH-334-MC-B	40.5	59.7	x		70°28′12.96″ N	24°17′0.66″ E
2	CAGE-22-1-HH-334-MC-C	36.5	59.7		x	70°28′12.96″ N	24°17′0.66″ E
3	CAGE-22-1-HH-343-GC	60.0	59.8		x	70°28′4.80″ N	24°17′21.77″ E
4	CAGE-22-1-HH-346-BC-C	13.0	47.7	x		70°27′28.68″ N	24°17′23.58″ E
5	CAGE-22-1-HH-347-BC-A	30.5	51.6		x	70°27′48.36″ N	24°16′16.3″8 E
6	CAGE-22-1-HH-352-BC-B	22.0	59.3	x		70°28′4.50″ N	24°17′3.00″ E
7	CAGE-22-1-HH-357-BC	11.0	69.7		x	70°29′25.44″ N	24°15′58.80″ E

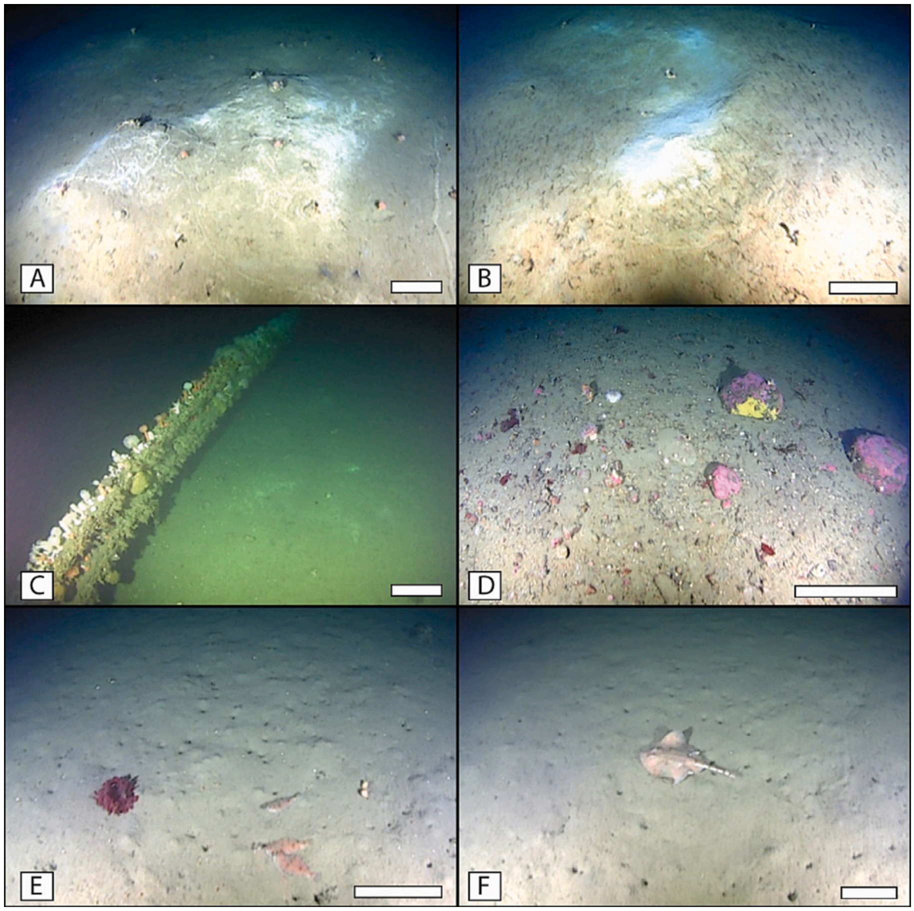

Fig. 2. Main seafloor characteristics of the mine tailings disposal area (A, B), the old pipeline colonized by fauna (C), sill (D) and the outer fjord (E, F) in the research area. Image sequences were converted from TowCam video recordings. Scale bar = 20 cm.

Gravity core 3 shows the lithostratigraphy of the uppermost 63 cm in the tailing area (Fig. 3). Sediment texture information was estimated by hand, based on stickiness, flexibility and roughness. The surface sediment comprises silty-fine sand with few shell fragments, passing at 11 cm to tailing interval. The tailing material is light olive brown in color, fine grained (clay size) and homogeneous in texture. It is barren of shells and very sticky. This interval is approximatively 3.5 cm thick and overlies a dark grey layer, very sticky and made of muddy silt. This layer might also be affected by tailing material. The mine tailing interval was found at depths between 5 and 13 cm in cores from the same area in previous studies (Sternal et al., 2017; Mun et al., 2020b). The pristine marine sediment in the lower part of the core from ~25 cm downwards is dark grey medium-fine sand containing some shell fragments and plane-parallel lamination.

4.2. *Pore water geochemistry*

4.2.1. *Redox conditions and sediment biogeochemical zonation*

Determining the redox conditions in the sediments is essential to reconstruct metal speciation and mobility as well as biogeochemical processes. The redox zonation of marine sediments is dictated by conditions in the bottom waters, the magnitude and rates of organic matter mineralization and biological activity reworking the surface sediment (Jørgensen and Kasten, 2006). In general, the uppermost sediment layer is rich in oxygen diffusing from the overlying water column. Microorganisms rapidly consume the oxygen as an electron acceptor in the process of organic matter oxidation. Below the oxic zone, less thermodynamically favourable compounds (yielding lower energy gain for the microbes) are consumed in the following order: nitrate (NO_3^-), manganese (oxy)hydroxides, iron (oxy)hydroxides and sulfate (SO_4^{2-}) (Jørgensen and Kasten, 2006). This succession corresponds to a steep decrease in redox potential and the pH, initially steeply decreasing within the oxic zone as a result of aerobic respiration and reoxidation of reduced species, tends to increase with depth due to anaerobic alkalinity production (Silburn et al., 2017).

The full results of the geochemical analyses of pore water are listed in Supplementary Information 1. In the study area, bottom waters have a pH of ~8 whereas pore water values measured in cores 1, 4 and 6 range from 7.2 to 7.8, from 7.4 to 8 and from 7.4 to 7.9 (Fig. 4). The pH profiles show a steep decrease in the uppermost 3–4 cm (Fig. 4) and rather stable values downcore. In core 6, the interval 6–11 cm shows the lowest pH values. The redox potential in the cores decreases from 226 mV in the well-oxygenated bottom waters, to negative values as low as −170.5 mV (33 cm, core 1) indicative of more reducing conditions downcore (Fig. 4). The Eh values become negative starting at 3 cm in all the cores. The mean Mn and Fe concentrations in bottom water are 0.045 ± 0.004 μM (2SD; $N = 2$) and 0.46 ± 0.10 μM (2SD; N = 2), respectively. Core 1, from the tailing area covered with tubeworms, shows Mn and Fe concentrations ranging from 0.12 μM to 0.97 μM and from 0.53 μM to 37.92 μM, respectively (Fig. 4). Core 4, close to the river delta, shows Mn and Fe concentration values from 0.43 μM to 1.56 μM and from 2.62 μM to 49.98 μM, respectively. Core 6, from the tailing area with no tubeworms, shows Mn values from 0.52 μM to 7.52 μM and Fe values from 3.59 μM to 87.65 μM. Mn concentration profiles in cores 4 and 6 display an overall increasing trend downcore and two humps centred at 2 and 4 cm depth, respectively, in correspondence with a change in gradients from high to low. These intervals correspond to production rates of 0.12 nmol cm^{-3} s^{-1} and 0.01 nmol cm^{-3} s^{-1}. The shape of the Mn profile in core 1

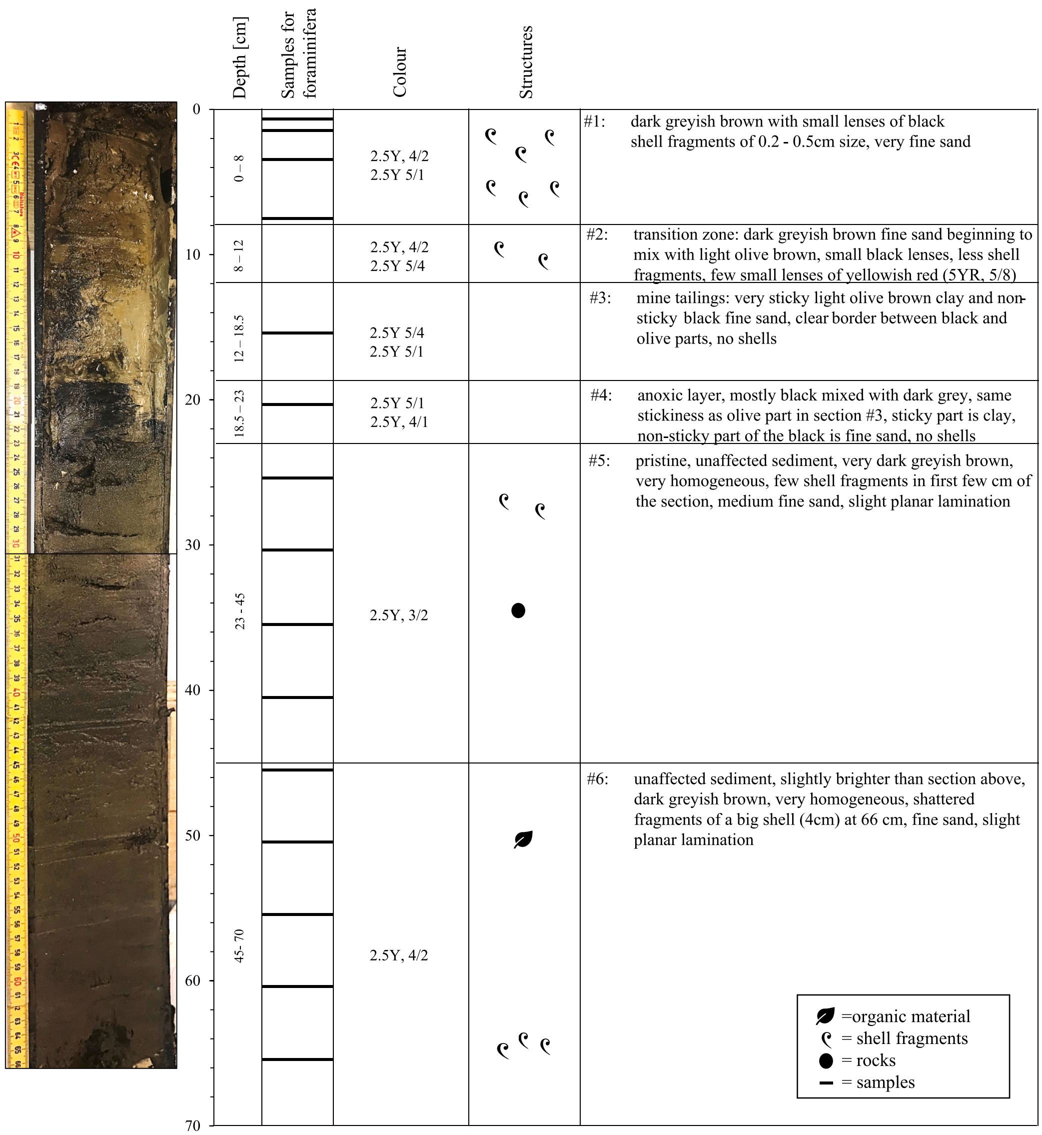

Fig. 3. Core description of gravity core 3, including sampling points (black lines) for benthic foraminifera. Color codes area based on the Munsell Soil Color Chart.

is different, being characterized by a well-developed broad peak from 0 cm to 15 cm depth, followed by near zero values downcore (Fig. 2). The Mn production rates are similar to core 6, with a value of 0.02 nmol cm^{-3} s^{-1}. The Fe profiles show similar trends to Mn (Fig. 4). Evident bulges are observed at ~3 cm in core 4 and ~5 cm in core 6, followed by a new quasi-linear increase downcore. Those intervals host production rates of 13 nmol cm^{-3} s^{-1} and 4 nmol cm^{-3} s^{-1}. Iron in core 1 behaves similarly to Mn, with the highest values found between 1 cm and 13 cm and a very low production rate < 0.5 nmol cm^{-3} s^{-1}. Sulfate concentrations range from 25.6 to 27.8 mM (Fig. 2) (seawater concentration is ~28 mM). The profile shapes are characterized by gentle gradients and display a slow decreasing trend starting below the manganese and iron profile bulges in all the three cores. Sulfate reduction rate is −8 nmol cm^{-3} s^{-1} in core 1, whereas it is zero in core 4. Positive values are artifacts. We measured low methane concentrations at the bottom of cores 2, 3 and 5, ranging between 1.0 μM and 5.9 μM.

The Fe and Mn profiles showed typical early diagenesis patterns (i.e. post-depositional processes taking place in aquatic sediments), where Fe and Mn oxides function as electron acceptors for various enzymatic pathways in the suboxic zone (Haese, 2006). The reduction of Fe and Mn (hydr)oxides in the suboxic zone causes the soluble divalent forms Fe^{2+} and Mn^{2+} to accumulate in the interstitial water, generating a bulge in the pore water depth-profile (Schulz, 2006). Since the reduction of Mn is thermodynamically more favourable, the peak in dissolved Mn typically appears above the one of dissolved Fe (Haese, 2006). This is visible in Fig. 4, where Mn bulges are located immediately above the Fe ones. By definition, the suboxic zone is followed by the anoxic zone of sulfate reduction (Jørgensen and Kasten, 2006). Consistent with the expected zonation, in our cores, we observe sulfate starting to decline below the Mn and Fe reduction zones. This redox zonation is accompanied by a decline in the redox potential from 226 mV in the bottom water down to negative values starting at 3 cm below the seafloor (Fig. 4). We measured low methane concentrations in the headspace gas from the bottom of cores 1, 3 and 5, corresponding to 1.0 μM, 4.2 μM and 5.9 μM, respectively, thus confirming that the coring retreat did not intercept the methanogenic zone which is by definition located below the sulfate reduction zone.

Unexpectedly, dissolved Fe and Mn concentrations increase again with depth at Sites 4 (reference site) and 6 (mine tailings affected site). This coincides with elevated concentrations measured in the sediment of the resembling cores (Huljek, pers. comm.) and suggests a production zone of Fe and Mn within the anoxic layer at approximately 15–20 cm depth. The unstable phase is most likely of anthropogenic origin and approximately correlates with the depth of the mine tailings layer

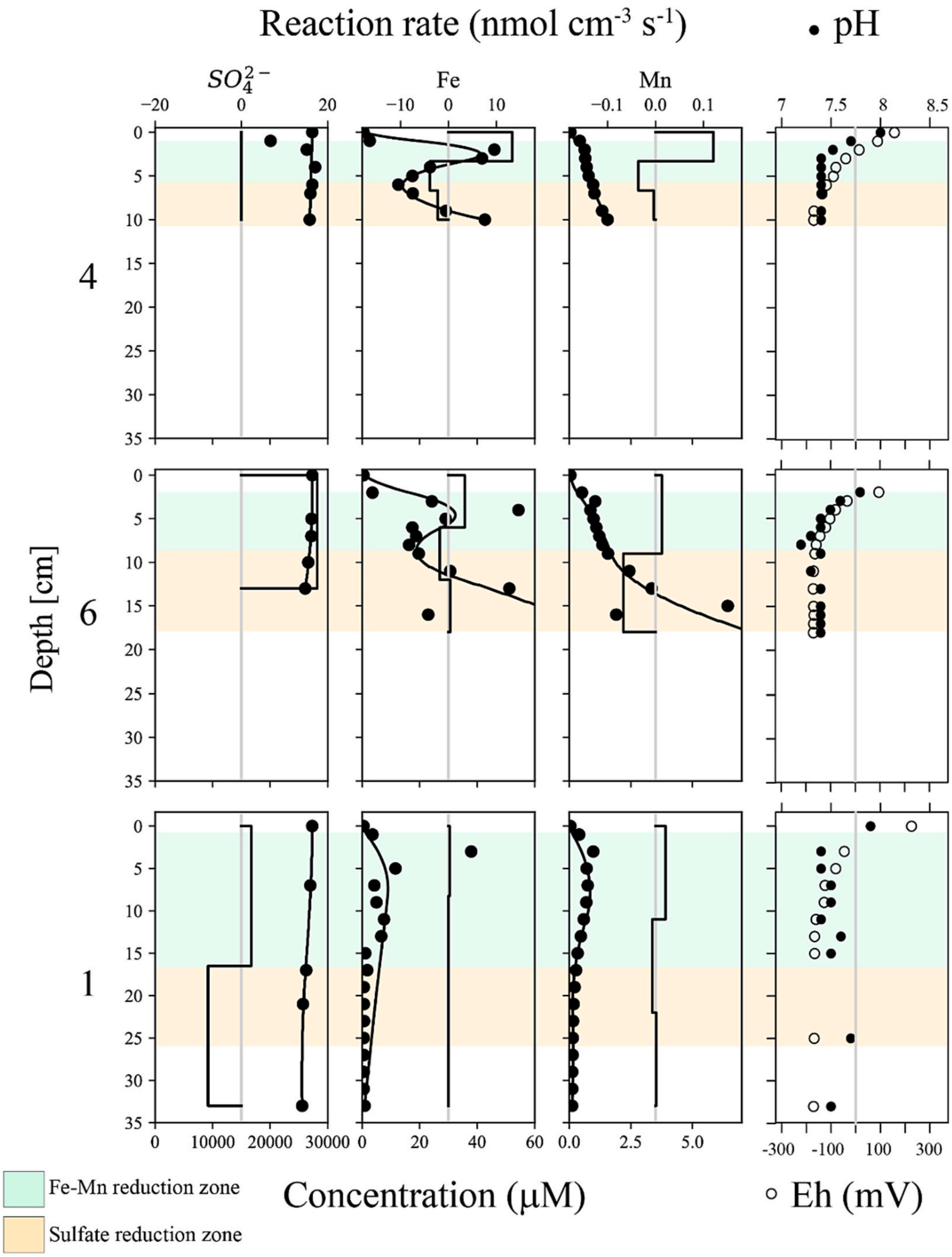

Fig. 4. Pore water-concentration profiles of sulfate, Fe and Mn with measured datapoints (dots), best fitting concentration profile (curved lines) and modelled reaction rates (consumption and production when negative and positive, respectively) (straight lines) in core 4 (reference area, inner fjord), 6 (tailings area) and 1 (tailings area, tube worm habitat). Two values of Fe at the bottom of core 6 exceed 60 μM and are reported in the Supplementary Information. Eh and pH values measured onboard are also reported. Background colors refers to interpreted redox zonation.

observed as a fine-grained sediment slurry between 12 and 23 cm depth in core 3 (Fig. 3). This phenomenon was not only observed at the mine tailings affected site (core 6), but also at reference core 4, which is located close to the river delta in the inner fjord. The origin of the deeper reactive sediment layer remains unclear. The reason why this deeper increase in Fe and Mn was not observed at the mine tailings-affected core 1 might be due to this site being influenced by bioturbation (e.g. Schaanning et al., 2019), as a dense colonization by polychaetes was observed during the video imaging survey of the research area.

4.2.2. Trace metals geochemistry

Concentrations of dissolved Cu in bottom water is 0.037 ± 0.005 μM (2SD; $N = 2$) whereas in cores 1, 4 and 6 show ranges of 0.02–0.07 μM, 0.02–0.11 μM and 0.03–0.09 μM (Fig. 5; Supplementary Information 1). Pore waters extracted from the reference core (the delta area out of the historical STD, core 4) show the Cu concentrations between 0.02 μM and 0.11 μM falling mostly within the range defined for "good" water quality according to the classification proposed by Miljødirektoratet for coastal waters (Guideline M-608, 2016). The sample extracted from 1 cm depth, with 0.11 μM, fits in the category of "very bad" water quality. The Cu content in this core reflects the fluvial inputs from the drainage basis geochemically controlled by Cu-rich volcano-sedimentary lithologies of RTW (Mun et al., 2020a). Copper in pore waters extracted from this core shows a statistically significant positive correlation with a range of chalcophile elements such as Cd ($r = 0.77$), Ni ($r = 0.90$), Sb ($r = 0.94$), Se ($r = 0.83$), and Sn ($r = 0.93$), reflecting their common source in sulfide mineralization oxidized under environmental conditions. In contrast, Cu shows a negative correlation with lithophile elements including Cr ($r = -0.63$), Li (r = −0,75), Rb (r = −0,97) and Sr (r = −0,94). Pore waters extracted from core 1 (historical tailing site inhabited by tubeworms) show the Cu concentration in the range between 0.02 and 0.07 μM, falling predominantly in the category of coastal waters of "good" quality (Guideline M-608, 2016). Core 6 shows the highest values and a bulge at ~5 cm depth followed by an increase toward the bottom of the core. The water quality status in this core according to national schemes is "bad" to "very bad".

The Ni concentration in bottom water is 0.01 ± 0.01 μM (2SD; $N = 2$), whereas in cores 1, 4 and 6 shows ranges of 0.005–0.01 μM, 0.003–0.02 μM and 0.003–0.03 μM (Fig. 5). The core near the delta shows a slightly decreasing trend from the top sediment layer to the bottom of the core. Most values fall in the "background" water quality category for coastal waters, whereas samples in the top 2 cm and at 5 cm are slightly higher. The Ni concentrations in Core 6 steadily decrease from the seafloor to a depth of ~10 cm and increase again downcore. The water quality is "background" in the middle portion of the core and "good" at the top and bottom. Core 1 shows a decrease in the concentration in the uppermost few centimetres and a bulge from 11 to 17 cm.

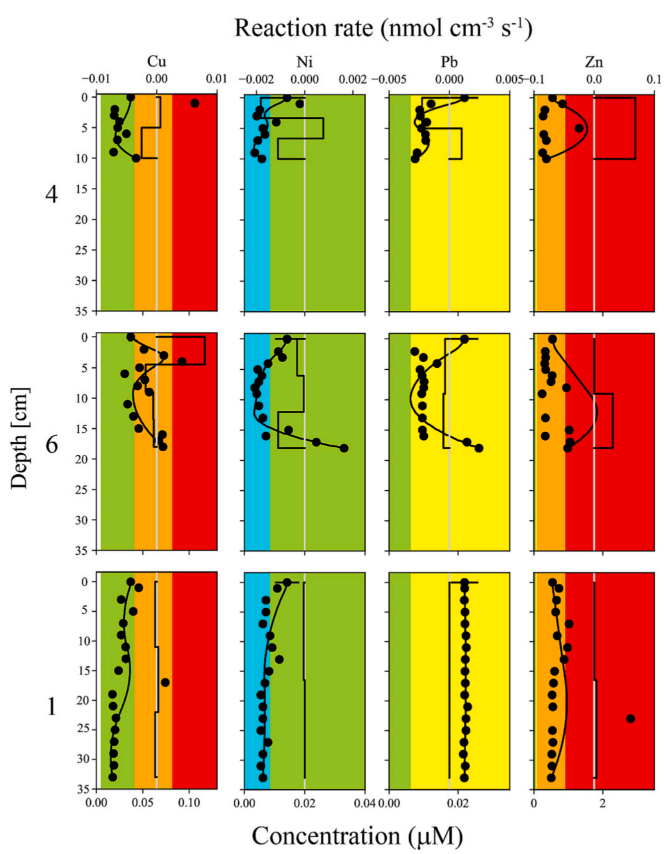

Fig. 5. Pore water-depth profiles of Cu, Ni, Pb and Zn with measured datapoints (dots), best fitting concentration profile (curved lines) and modelled consumption and production profile (straight lines) at Sites 4 (reference area, inner fjord), 6 (tailings area) and 1 (tailings area, tube worm habitat). Background colors refer to the Norwegian environmental quality classification of metal contaminants in coastal waters defined by the Norwegian pollution control authority Miljødirektoratet (Guideline M-608, 2016): blue = "background"; yellow="moderate"; orange = "bad"; red = "very bad".

The most of values can be classified as "background" quality whereas the values at the top and in correspondence of the bulge shape are "good".

The Pb concentration in the bottom water sample is 0.022 ± 0.001 μM (2SD; N = 2), whereas in the cores it ranges between 0.022 μM and 0.02 μM in core 1, from 0.008 μM to 0.01 μM in core 4 and from 0.007 μM to 0.03 μM in core 6 (Fig. 5). Shapes of the concentration profiles in cores 4 and 6 are similar to the profiles of Ni, whereas core 1 has a flat profile (no gradient) with values stable at around 0.02 μM. All cores have pore waters classifiable as "moderate" quality status in regards of Pb.

Zinc in bottom water has a concentration of 0.54 ± 0.03 μM (2SD; N = 2); cores 1, 4 and 6 shows ranges of 0.51–2.81 μM, 0.26–5.54 μM and 0.24–7.45 μM, respectively (Fig. 5). The concentration profile is rather flat in core 4 except for an elevated value at 5 cm. The profile in core 6 is scattered below 5 cm but shows an overall increase in concentrations below that depth. Core 1 shows a rather flat profile with relatively higher values found between 7 cm and 13 cm and at 23 cm. Overall water quality in the three cores is classified as "bad" to "very bad" following to national Zn thresholds for coastal waters. It is worth mentioning that the categories defined by Miljødirektoratet (Guideline M-608, 2016) refer to coastal waters and do not consider the influence of local fjord geology, which can greatly impact the definition of natural background.

The heavy metals content of the natural (background) sediments at Repparfjord is controlled by fluvial input of Repparfjordelva that drains volcano-sedimentary lithologies of the Repparfjord Tectonic Window, including metamorphosed mafic volcanic (pillow lavas, breccias and tuff) and carbonate-siliciclastic meta-sediments (dolomitic marbles, metasandstones, metasiltstones). The latter formations host numerous Cu occurrences composed of sulfides such as chalcopyrite ($CuFeS_2$), bornite (Cu_5FeS_4), chalcocite (Cu_2S) and covellite (CuS), with minor amounts of pyrite (FeS_2) and sphalerite ((Zn, Fe)S) (Torgersen et al., 2016; Mun et al., 2020a). Previous studies on bulk sediment geochemistry found that the tailing-impacted layers are remarkably enriched in Cu and Ni compared to the background sediments (Sternal et al., 2017; Mun et al., 2020b). Other potentially toxic metals such as As, Cd, Co, Pb, and Zn were found in low amounts, similar to the sediments carried by the Repparfjordelva. Other minerals found in the natural Repparfjord sediments include quartz (SiO_2), plagioclase ($(Na,Ca)(Si,Al)_4O_8$), and micas. These phases are rather stable during early diagenesis.

From the tailing area, core 6 showed indication of Cu release from the sediment into pore water in two stratigraphic intervals: one narrow interval characterized by a bulge in concentration profile centred at around 5 cm and another interval located below 18 cm. The uppermost signal strikingly matches the vertical distribution and average sediment thickness of the tailing deposits in core 3 and other cores from previous studies (Sternal et al., 2017; Mun et al., 2020b). The Repparfjord sediment contains high amounts of Cu associated with both the tailing material and the background sediment derived from weathering of bedrock lithologies. As demonstrated in a previous study by Mun et al. (2020b), Cu in the tailing material is mainly associated with the acid-soluble fraction (carbonates) (45 %) and reducible (Fe—Mn oxyhydroxides), exchangeable and oxidizable (bound to organic matter and sulfides) fractions, each one containing 15 % of the total Cu. The residual fraction, e.g. silicates, makes up the 10 % of the total. The latter is stable at low T diagenetic conditions occurring in the uppermost tens of centimetres of sediments, whereas all the other fractions are potentially altered and able to release Cu into pore waters at appropriate pH-Eh conditions. In the natural sediments, Cu is mainly hosted in residual phases, therefore presenting low mobility. Scanning electron microscopy analyses have shown that the sulfides in Repparfjord sediments are generally well preserved except for sphalerite which shows occasionally traces of oxidation (Mun et al., 2020b). This might partially explain the high background zinc concentrations found in pore waters in all the cores as related to the Repparfjord Tectonic Window lithologies. Copper appears to be mobilized in the suboxic zone, with concentration trends matching the Fe and Mn profiles in core 6 and partially distributed within the sulfate reduction zone. Iron in the sediment also shows the highest contents in the same interval, whereas sulfur is only in trace amounts (Huljek, pers. comm.). In order to interpret the source of Cu in the pore waters we conducted geochemical modelling using PHREEQC software. We used the measured Eh-Ph and elemental compositions in pore water samples from cores 4 and 6 as input solutions to calculate Cu speciation and the stability of Cu-bearing solid phases at different sediment depths. In core 4, chalcopyrite, covellite, chalcocite and sphalerite (the main ore minerals in the Nussir and Ullverygen mines) are all unstable (tend to dissolve) in the interval 0–5 cm and become stable downcore (Supplementary Information 2). Specifically, chalcocite is stable at 5 cm, chalcopyrite and covellite at 6 cm and sphalerite is stable at 7 cm. Pyrite is stable from 7 cm downwards. In core 6, chalcocite becomes stable at 4 cm, while chalcopyrite and covellite are stable from 6 cm downward. Sphalerite and pyrite are stable from 7 cm. The tailing material is partially exposed at the seafloor in many areas of the inner fjord as seen during the towed-camera dives. Based on our models and analytical results, we believe that the patchy distribution of the exposed tailings might correspond to loci of higher metal leaching directly into the water column. Capping the tailings with natural clays as suggested in other studies (Schaanning et al., 2019; Trannum et al., 2023) can bury the tailings below the "reactive zone" of the sediment, keeping the material stable. Overall, the models established that the source of Cu is in sulfides from the ore materials, and their dissolution in the uppermost (~0–5 cm) interval, but fail to describe the signals in other elemental trends which remain unclear. The highest Cu leaching rate in the tailing interval in core 6 corresponds to 0.008 nmol cm^{-3} s^{-1}. Leaching of Ni, Zn and Pb rates up to 0.0008, 0.001 and 0.08 nmol cm^{-3} s^{-1}, respectively, have been attributed to weathering of natural bedrock lithologies. The sediment accumulation rates in the inner fjord determined by Sternal et al. (2017) are in the range 1.5–4.3 mm/yr, ascribing the sediments in the lowest part of the pore water Cu bulge in core 6 (10 cm) to ~1955–1999 CE and the sediments at the bottom of the core showing Cu increasing trend (20 cm) to <1889–1975 CE. Based on these values, both Cu enrichments in the core 6 would be consistent with leaching of tailing material disposed from the 1972. In fact, the bulk sediment geochemistry of core 6 shows an overall enrichment in Cu compared to core 4 (delta site), and two pronounced peaks at 9 and 20 cm (Huljek, pers. comm.) which matches the pore water observations. Nickel is bonded to mafic silicate minerals such as olivine and pyroxene deriving from the bedrock, whereas Pb and Zn can be also associated with the ore mineral assemblages. Pore water modelling revealed some leaching of these elements at rates up to 0.0008, 0.001 and 0.08 nmol cm^{-3} s^{-1}, respectively. Their sharp increase in concentrations at the bottom of core 6 suggest a deeper source for these metals being released into pore waters which is tentatively attributed to weathering of mineral assemblages from natural bedrock lithologies and requires further investigation.

4.2.3. Benthic fluxes

Natural sediments act as a source or sink of trace elements to or from the water column. Hereby, the first zone in the sediments pore water, which reached depths of up to 6 cm in the research area, determines the gradient across the sediment-water interface. Typically, concentrations of dissolved trace metals within the pore water exceed those in the bottom water, resulting in net benthic fluxes of dissolved metals to the water column (Santos-Echeandia et al., 2009). For that reason, applying the M-608 classification scheme for coastal waters to pore waters might lead to overestimations of the contamination level. There is a need of further studies to address the relationships between benthic ecosystems and metal concentration and bioavailability in sediment pore waters in both natural and contamined areas, to be able to set baseline values and environmental thresholds.

The benthic fluxes modelled with the PROFILE software significantly differed from those calculated based on the best linear fit since the software tends to smoothen peaks observed in the oxic and suboxic zone and is less sensitive to outlier values. We, therefore, assume that the latter ones reflect the actual conditions more accurately. Therefore, only those were considered for the discussion and comparison to other polluted sites.

Fluxes of Fe and Mn into the water column were observed in every core, as Fe^{2+} and Mn^{2+} diffuse upwards from the suboxic zone. For Ni, negative fluxes from the water column into the sediment, were observed due to a high average concentration of 0.02 μM in the bottom water (Table 3). These can stem either from input via river discharge, as Ni is naturally abundant within mafic rocks which predominantly compose the bedrocks in the research area (Mun et al., 2020a) or from anthropogenic activities, such as wastewater discharge and boating activities.

Fluxes of Pb from the water column into the sediments pore waters were observed (Table 3), most likely due to Pb being naturally enriched in the bedrock and therefore brought into the coastal water by weathering of the surrounding rocks (Mun et al., 2020a). While Zn was generally present in high concentrations in the research area, positive fluxes from the sediment pore water into the water column were only observed in cores 1 and 4 (Table 3).

Fluxes of dissolved Cu into the water column were observed throughout all sites confirming the sediment as a net source of copper for the fjord system, while the highest fluxes of 79.0 nmol cm^{-2} yr^{-1} occurred at reference core 4, followed by core 6 with a flux of 15.7 nmol cm^{-2} yr^{-1} and core 1 with a flux of 9.9 nmol cm^{-2} yr^{-1}. However, Cu concentrations were generally lower at the reference site than at the other two sites, and the high benthic flux is mainly due to a very high concentration of Cu measured at a depth of 1 cm. Since this peak was not in accordance with concentrations of Cu measured within the sediment (Huljek, pers. comm.), it could potentially be an outlier. Without considering the high concentration measured at 1 cm depth, the resulting flux of Cu in core 4 would be −7.2 nmol cm^{-2} yr^{-1}.

Modelled fluxes of trace metals in estuarine areas impacted by STD are not frequently reported. However, several studies determined trace metal benthic fluxes in natural and contaminated sediments based on pore water or DGT (diffusive gradient in thin films) probing profiles for various contaminated ecosystems. A selection is given in Table 3. Trace metal fluxes across the sediment-water interface of the two mine tailings affected sampling cores 1 and 6 lay between fluxes calculated for low contaminated coastal and estuarine ecosystems (Vigo Ria (ES), Dyngadjupet (NO) and Gullmarsfjord (SE)) and for polluted ecosystems (Jøssingfjord (NO), Thau Lagoon (FR) and Ansedonia Bay (IT)). It should, however, be noted that those sites vary geochemically and comparing trace metal fluxes between different estuarine areas is

Table 3
Exchange fluxes of dissolved trace metals at the sediment water interface in the Repparfjord and other estuarine areas. Fluxes are given in nmol cm^{-2} yr^{-1}.

location	Cu	Ni	Pb	Zn	Reference
Repparfjord 4 Ref (NO)	79.0	−4.84	−6.2	322.9	this study
Repparfjord 6 (NO)	15.7	−1.4	−44.6	−43.8	this study
Repparfjord 1 (NO)	9.9	−1.2	0.1	209.1	this study
Vigo Ria (ES)	−0.02-1.1	0.2–1.7	−0.1-0.03	−0.37-0.9	(Santos-Echeandia et al., 2009)
Jøssingfjord (NO)	213.1	27.6			(Schaanning et al., 2019)
Dyngadjupet (NO)	174.0	37.1			(Schaanning et al., 2019)
Gullmarsfjord (SE)	0.9–4.3	−2.2-14.6		51.1–69.4	(Westerlund et al., 1986)
Thau Lagoon (FR)	−416-1124		−18.3-153.3		(Point et al., 2007)
Ansedonia Bay (IT)	−0.7		−3.7-(−4.14)	54.9–254.5	(Ciceri et al., 1992)

therefore difficult.

4.3. Benthic foraminiferal community – past and present status

Twenty-seven benthic foraminiferal species of eight different genera were recorded (see supplementary information 3), of which 16 included living individuals. The average absolute abundance of foraminifera throughout all samples was 13.1 ± 10.3 ind.g^{-1} for the living specimens and 2316 ± 1095 ind.g^{-1} for the dead ones. While the outer fjord displayed the lowest absolute numbers of species, the other sites lay relatively close to each other.

4.3.1. Comparison of three assemblage clusters

The disposal of mine tailings into the Repparfjord resulted in significant changes in the assemblage composition of benthic foraminifera. Fig. 6 shows the relative distribution of foraminifera sorted by species, sampling site and depth and displays differences in the composition of the communities between the sampling sites that are supported by the results of the NMDS analysis shown in Fig. 7. The NMDS revealed three main clusters, each including samples with similar species and shell type: (1) The samples taken from core 3 at depths >21 cm, thus located below the mine tailings layer. (2) Samples from the upper part of the same core and all samples from the tailings impacted cores 2 and 5. (3) Samples from the outer fjord at core 7.

About 80–140 years of sedimentary record are preserved in the lower part of core 3 (Fig. 3) assuming a sedimentation rate of 0.28–0.5 cm yr^{-1} based on previous studies (Andersson et al., 2018; Sternal et al., 2017). During this period, the occurring species remain the same, but their relative distribution changed over time, probably indicating a natural decline in bottom water oxygen levels from 1810 to 1870 to 1970. This is revealed by the relatively high abundance of calcareous species at the bottom of the core that declines toward the younger sediment layers together with a relative increase of agglutinated species (Elberling et al., 2003).

The foraminiferal assemblage of the lower part of core 3, dominated by *Adercotryma glomeratum*, *Buccella* spp., *Lobatula lobatula* and *Cribroelphidium excavatum*, disappeared entirely during the disposal period, due to the hostile conditions that prevailed in the mine tailings. The large volume of fine-grained material smothered the habitat and all associated sessile biota, destroying the faunal community during disposal (Ramirez-Llodra et al., 2015). After the discharge had been

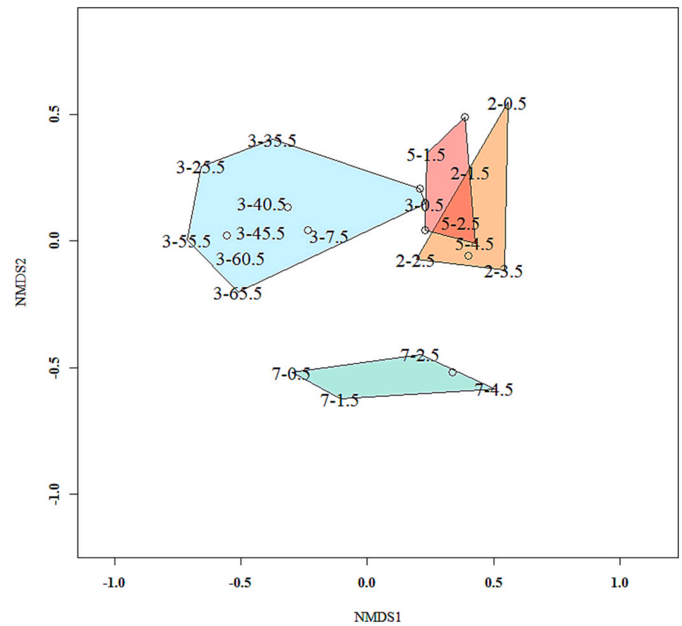

Fig. 7. NMDS plot of benthic foraminifera data in the Repparfjord. Numbers indicate the sampling site and sampled depth, while colored regions encompass the different sampling sites: core 2 – orange color – tailings area; core 3 – blue color – tailings area; core 5 – red color – tailing area; core 7 – green color – outer fjord.

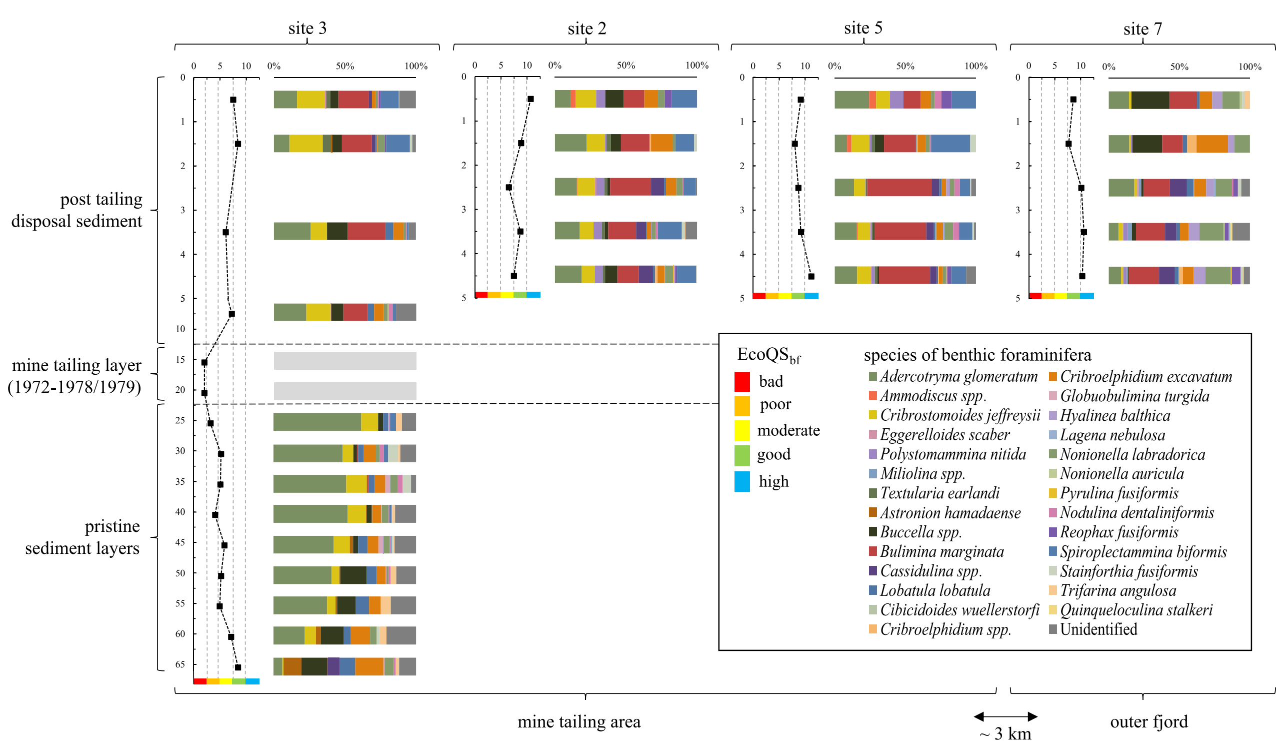

Fig. 6. EcoQS$_{bf}$ – depth profiles and relative distribution of benthic foraminiferal species in cores 2, 3, 5 (mine tailings impacted areas, inner fjord) and 7 (reference area, outer fjord). Y-axis indicates sampled depth. The diversity is expressed as exponential of the bias corrected Shannon-Wiener index, while the colors reflect the corresponding environmental quality status (EcoQS$_{bf}$) defined by Bouchet et al. (2012). The historical mine tailings are located at approximately 12–23 cm depth.

stopped in 1979, a new benthic community began to be established. Presumably, benthic foraminifera living in the surrounding area colonized the mine tailings affected area. However, the immigrant assemblage exhibits a different composition than the natural assemblage prior to the tailings discharge. In addition to *A. glomeratum*, and *Buccella* spp., the species *B. marginata*, *C. jeffreysii* and *S. biformis*, all of which were absent prior to the mining activity, are now dominating in the community, while the presence of *Trifarina angulosa*, *Astronion hamadaense* and *Globuobulimina turgida* diminished considerably reaching the point of nearly disappearing from the assemblage almost completely. The critical difference between the dominant foraminiferal assemblage downcore (i.e. the first cluster, and the second cluster) and sediments above the mine tailings layer is the relatively higher abundance of opportunistic and stress tolerant species like *B. marginata* and *S. biformis* in the latter (Dijkstra et al., 2017; Mojtahid et al., 2006; Polovodova Asteman et al., 2015). As shown by the EcoQS values (Fig. 6, Table 4), the overall diversity and density of foraminiferal species recovered to historical values but there was a significant change in species composition from prior to the mine tailings disposal to afterwards.

Agglutinated taxa are present in relatively high abundances only in sediments deposited prior to the mining activity. Agglutinated taxa exhibit a higher resistance to dissolution compared to calcareous, and their absence, together with the results from pore water data, could suggest vertical leaching and dissolution of calcareous taxa, as pH values in Cores 6 and 1 tend to decrease throughout the core. Comparing pre- with post-deposition, where agglutinated and calcareous foraminifera accounted for approximately half of the assemblage each, stands in contrast to other observations from contaminated coastal sites, where agglutinated taxa dominated at highly impacted Sites (Alve, 1991; Polovodova Asteman et al., 2015). Other studies, however, reported a

Table 4
Environmental classification of cores 2, 3, 5 and 7 based on foraminiferal investigations. Number of counted specimens per sample (n), number of species in the sample (S), total standardized abundance of living and dead foraminifera in individuals per gram bulk dry sediment (# g^{-1}). ExpH′ reflects the diversity of benthic foraminiferal assemblages, and F-AMBI weights the species regarding their sensitivity to organic matter. In contrast, the colors reflect the EcoQS classes according to Bouchet et al. (2012) and Bouchet et al. (2021). The ecological quality ratio (EQR) was determined for core 3 based on the highest ExpH′ value at 65.5. cm depth. FAI [%] represents the relative foraminiferal abnormality index.

site		core depth [cm]	n	S	# g^{-1}	ExpH′	EQR	F-AMBI	FAI [%]
2	inner fjord,	0.5	55	10	n.a.	10.69	n.a.	2.1	7.27
	tailings area,	1.5	128	14	n.a.	8.81	n.a.	2.8	3.91
	tube worm habitat	2.5	303	14	1222	6.50	n.a.	2.4	3.30
		3.5	315	20	1456	8.69	n.a.	2.4	2.54
		4.5	305	16	1734	7.46	n.a.	2.5	2.95
3	inner fjord,	0.5	300	13	740	7.52	0.88	3.3	7.33
	tailings area	1.5	119	13	590	8.46	0.99	3.7	9.00
		3.5	158	9	1350	6.11	0.71	2.8	6.96
		7.5	176	11	831	7.37	0.86	2.7	3.98
		15.5	4	2	11	2.09	0.24	n.a.	n.a.
		20.5	4	2	15	2.09	0.24	n.a.	n.a.
		25.5	143	8	1895	3.27	0.38	1.2	4.90
		30.5	200	11	1025	5.28	0.62	0.9	4.50
		35.5	200	11	450	5.16	0.60	1.2	4.00
		40.5	277	12	1522	4.13	0.48	1.4	7.67
		45.5	261	13	841	5.94	0.69	1.4	1.38
		50.5	145	10	338	5.28	0.62	1.8	9.66
		55.5	82	7	508	5.03	0.59	1.4	3.53
		60.5	87	9	500	7.24	0.84	2.2	1.15
		65.5	127	12	709	8.57	1.00	1.8	4.72
5	inner fjord,	0.5	63	10	n.a.	9.15	n.a.	2.5	6.35
	tailings area	1.5	103	11	n.a.	8.02	n.a.	2.1	8.74
		2.5	301	14	2034	8.68	n.a.	2.2	2.99
		3.5	301	15	1011	9.18	n.a.	2.1	3.32
		4.5	302	16	3657	11.16	n.a.	1.9	4.30
7	outer fjord,	0.5	61	11	n.a.	8.57	n.a.	1.5	3.13
	reference area	1.5	99	10	n.a.	7.67	n.a.	1.4	6.06
		2.5	315	16	523	10.11	n.a.	1.4	2.22
		3.5	315	17	228	10.60	n.a.	1.9	2.54
		4.5	315	17	331	10.29	n.a.	1.7	2.86

low presence of agglutinated taxa attributed to the release of oil and tar due to heavy shipping traffic in the port of Hammerfest (Dijkstra et al., 2017). Interestingly, agglutinated taxa are also less abundant in sediments influenced by natural methane seepage, suggesting that this group of foraminifera does not tolerate the geochemical conditions at seeps (Dessandier et al., 2019).

The palaeosedimentary record below the mine tailings affected layers is more suitable for establishing a habitat baseline than the presumably pristine foraminiferal assemblage in the outer fjord. The community composition and the total abundance of benthic foraminifera in the outer fjord differed from the previously described clusters. While *A. glomeratum* and *B. marginata* were present in the same quantities as in the inner fjord, *Cassidulina* spp. and *Nonionella labradorica* were unique to this location. Consequently, the relative abundance of calcareous taxa was highest in this area, resembling good living conditions and a high oxygen level in the bottom water. In contrast to the inner fjord, the absolute abundance of benthic foraminifera was considerably lower in the outer fjord. As the community structure of benthic foraminifera depends on various factors, it would be highly speculative to attribute the differences in this cluster to the presence or absence of mine tailings. It is more likely these differences stem from discrepancies in the geographical setting of the inner and outer fjord, i.e. a different water temperature and salinity, different nutrient availability and greater water depth. Therefore, retroactively establishing reference conditions from pristine sites is difficult. Due to the high spatial variability of foraminifera, a pristine reference Site would need very similar ecological characteristics to the impacted site. However, drawing from the proper reference conditions is crucial to assess the ecological quality. The palaeosedimentary record from prior to the mine tailings discharge can provide such a reference condition.

4.3.2. Foraminiferal diversity and EcoQS

Benthic foraminiferal assemblages, even if composed by completely different taxa, have recovered their diversity and absolute abundance within approximately 14 years (assuming a sedimentation rate of 0.28 cm yr^{-1} based on 12 cm of sediment above the mine tailing interval in core 3) after 7 years of mine tailings disposal. The ecological status (Fig. 6) based on fossil benthic foraminifera and the classification scheme from Bouchet et al. (2012) ($EcoQS_{bf(H)}$) was moderate, good or high in the recent sediment of every location. If core 7 from the outer fjord does not represent a reliable reference condition, it is advantageous that using benthic foraminifera for environmental monitoring enables the reconstruction of the PalaeoEcoQS even though no data are available from this period. It is, therefore, assumed that $ExpH'_{bf}$ values around 8.57 from the lower part of the core 3 represent the baseline conditions for the inner fjord.

Interestingly, the $ExpH'_{bf}$ values are slightly lower, just below the mine tailing interval, than in recent sedimentary layers, i.e. some decades after the disposal of the mine tailings. In today's sediment, they range from 6.11 to 10.69. The relatively low EcoQS classes, even in the presumed pristine samples, lead to questioning the applicability of the threshold values for $ExpH'_{bf(H)}$ in Arctic ecosystems.

Since the classification is based on the response of benthic foraminifera to bottom water oxygen levels in Southern Norway (Bouchet et al., 2012), the threshold values might not be directly transferable to research sites located at higher latitudes. Dijkstra et al. (2017) were the first to draw attention to this problem, as their determined $EcoQS_{bf(H)}$ values in Hammerfest harbour reflected worse conditions than the EcoQS based on other variables, such as the concentration of pollutants and heavy metals, in the area. It is assumed that the natural diversity of benthic foraminiferal assemblages in Northern Norway is lower compared to more southern latitudes due to differences in water temperature and salinity (Armstrong and Brasier, 2005; Dijkstra et al., 2017). Therefore, the threshold values for benthic foraminiferal $EcoQS_{bf(H)}$ should be lower for higher latitudes. However, the $EcoQS_{bf(H)}$ reflects temporal trends and the benthic recovery and is still helpful for monitoring the ecological quality at high latitudes. One way to circumvent this issue is looking at the ecological quality ratio (EQR) instead, where recent EcoQS $_{bf(H)}$ values are set in relation to one from a presumably pristine environment. Table 4 shows the EQR values for gravity core 3, based on the assumption that the layer preceding the mining tailings with the highest EcoQSbf(H′) corresponds to the reference condition, which here is the deepest sediment layer (65.5 cm depth) with the highest $EcoQS_{bf(H)}$ (8.57). High EQR values between 0.71 and 0.99 in recent sediment layers support the previous assumption of a relatively fast recovery of the benthic foraminiferal ecosystem within approximately 14 years after the STD period. However, a comprehensive study is still needed to test the $EcoQS_{bf(H)}$ against other physical, chemical and organic stressors and adjust it for high latitude ecosystems.

A first conclusion regarding the impact of organic matter as one of these stressors can be drawn from looking at the $EcoQS_{bf(F\text{-}AMBI)}$ classes determined based on the F-AMBI index (Table 4). In general, the EcoQS classes based on the F-AMBI were higher (e.g. representing a better ecological quality) than the ones based on diversity. This supports the previous assumption, that the $EcoQS_{bf(H)}$ underestimates the true ecological quality. Assuming that the TOC in disposal area is low due to the mine tailings (Mun et al., 2020b), we expected higher F-AMBI values (i.e. a worse $EcoQS_{bf(F\text{-}AMBI)}$) in sediment layers directly influenced by the mine tailings. Indeed, all samples from layers where the sedimentation occurred after the tailing's disposal period, except the one from the outer fjord, display slightly higher F-AMBI values due to more species with a tolerance for low TOC in those layers. This is in accordance with the three assemblage clusters described above, where the recent ecosystem is dominated by more stress tolerant and opportunistic species.

4.3.3. Foraminiferal abnormality index

This study confirmed the findings of Melis and Covelli (2013), suggesting no significant effect of elevated trace metal concentrations on the development of morphological deformations of the shells of benthic foraminifera. The percentage of abnormalities showed no clear trend with depth or differences between the sampling Sites (Table 4). However, abnormalities were relatively common in the investigated area (> 1 %), reflecting a stressed environment in general (Coccioni and Frontalini, 2005). Therefore, it is suggested that the $EcoQS_{bf}$ is more suited than the FAI to monitor the status of an ecosystem impacted by STD.

5. Conclusions

The historical mine tailings in the Repparfjord are preserved as a fine-grained slurry in the sedimentary column within the disposal area and it is unclear whether they are releasing metals into the environment and their potential impact on ecosystems. We found that Cu, Ni, Pb, and Zn concentrations in some of the sediment pore water samples were elevated according to toxicity values defined by the Norwegian Pollution Control Authority. However, the threshold values in the national guidelines refer to coastal waters so they might overestimate the contamination level for pore waters which requires further studies. Lead and Zn enrichments indicate a strong control from bedrock lithologies dominated by a volcano-sedimentary complex, whereas the Cu anomalies are interpreted to reflect the ongoing dissolution of sulfides from the mine tailing. Copper enrichments are ascribed to tailing material disposed in the 1970s, that is, still releasing metals into the pore water system and out of the seafloor. Interestingly, metal leaching was also observed in natural sediments near the river delta, highlighting the need for a multidisciplinary approach to decipher natural vs anthropic signals.

The disposal of mine tailings into the fjord resulted in changes within the benthic foraminiferal community. The historical assemblage disappeared almost entirely during the disposal period. Foraminifera from the surrounding area colonized the disposal area in the 80s and recovered to the former diversity level within approximately 30 years. However, the present community is now dominated by more stress tolerant and opportunistic species that replaced less resilient species. Morphological deformations were observed in the investigated area but did not relate to trends in trace metal concentrations. The multidisciplinary approach used in our study can help improve the monitoring and mitigation of environmental challenges in the Arctic.

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CRediT authorship contribution statement

Marie Hoff: Writing – original draft, Visualization, Methodology, Investigation, Formal analysis. **Claudio Argentino:** Writing – review & editing, Visualization, Supervision, Methodology, Investigation, Conceptualization. **Laura Huljek:** Writing – review & editing, Investigation. **Željka Fiket:** Writing – review & editing, Investigation. **Ines Barrenechea Angeles:** Writing – review & editing, Supervision, Methodology, Investigation. **Sabina Strmic Palinkas:** Writing – review & editing, Supervision, Methodology, Investigation, Funding acquisition, Conceptualization. **Giuliana Panieri:** Writing – review & editing, Supervision, Methodology, Investigation, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Giuliana Panieri reports financial support was provided by Norges Forskiningsrad. Sabina Palinkas reports financial support was provided by Equinor ASA. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

All datasets are attached as supplementary materials.

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