Title: The impact of submarine copper mine tailing disposal from the 1970s on Repparfjorden, northern Norway

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Abstract:

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We investigate the state of sedimentological environment and contaminant status of Repparfjorden (N Norway) impacted by submarine disposal of mine tailings during the 1970's. Based on examination of the sedimentological and geochemical properties of seventeen sediment cores, the impact of tailings disposal is mainly restricted to the inner fjord where the discharge occurred. Sediment cores retrieved from the inner fjord contain layers of mine tailings up to 9-cm thick, 3-9 cm below the seafloor. Spreading of the tailing-related metal Cu and particles is limited to the inner fjord and to a 2 cm layer in one core from the outer fjord. Two interrelated factors, fjord morphology and sedimentation rate, controlled the distribution of contaminant-laden tailings in the fjord. The mobility of Cu from buried contaminated sediments to the sediment-water interface in the inner fjord indicates that benthic communities have been continuously exposed to elevated Cu concentrations for nearly four decades.

Keywords: fjord; submarine mine tailing disposal; contaminated sediments; grain-size distribution; heavy metals; sedimentation rate

1. Introduction

Coastal zones are of great ecological, economic and social importance (Martínez et al., 2007) and with increasing human activities they are particularly prone to anthropogenic pollution. Especially vulnerable are estuaries, forming a natural sink for (contaminated) sediments, that due to their dynamic regime may also act as sources of sediments, both seaward and up-estuary (Ridgway and Shimmield, 2002). There are different types of marine contaminants, originating from e.g. sewage effluents, agricultural run-offs, industrial effluents, oil spills, litter, etc. (e.g. Islam and Tanaka, 2004). Among them, although more marginal in a global scale, are mine tailings.

40 Mine tailings are a waste product resulting from the mechanical and chemical separation of minerals from geological 41 material retrieved during mining. They contain a mixture of milled ore (often of high residual metal concentrations), 42 water and sometimes process chemicals. The most common way of disposing and storing mine tailings is on-land (up to 43 99%) but an alternative, marine disposal, is practiced in Norway and Papua New Guinea, and at some locations in 44 Turkey, England, Greece, France, Chile and Indonesia (Vogt, 2013). Marine tailings disposal approaches are 45 differentiated based mainly on the depth of tailings discharge, an important factor from the perspective of potential 46 ecological and environmental impacts (for review see e.g. Ramirez-Llodra et al. 2015). However, recent legal 47 frameworks limit all types of marine disposal with exceptions under specific conditions, as for example described 48 within the London Protocol (IMO, 2016), a globally leading initiative to protect marine environment. 49

Post-discharge studies of historical submarine tailings disposal operations have identified impacts on the environment. They are primarily confined to the area of the disposal and may include physical and geochemical alteration of the bottom sediment, smothering of benthic organisms, reduction of marine biodiversity, risk of bioaccumulation of heavy metals in aquatic organisms (e.g. Burd, 2002; Elberling et al., 2003; Hughes et al., 2015; Josefson et al., 2008; Larsen et al., 2001; for review see also Ramirez-Llodra et al., 2015). Even though operators are required to establish practices to minimize the potential environmental consequences, some of the documented historical disposal activities have impacted a wider area than expected (e.g. Edinger, 2012; Perner et al., 2010).

58 Submarine tailings discharge has been carried out in Norwegian fjords for more than 100 years. There are at least 26 59 historical sites where relatively large discharges and/or discharges with the release of potentially toxic metals have 60 occurred (Kvassnes and Iversen, 2013). However, as underlined by Ramirez-Llodra et al. (2015), there are no reports 61 providing information on the exact composition of discharges, i.e. neither detailed chemistry and their changes, nor the

fate of these materials once they enter the marine environment. This gap of knowledge is of particular significance since fjords are estuaries with characteristic morphological features, circulation and stratification patterns. The presence of basin(s) separated by sill(s) make them depocenters for (contaminated) sediments, thus rendering them particularly susceptible to pollution impacts.

3 Various analytical tools are used to map the historical contaminations of the marine environment (e.g. estuaries) 4 recorded in sediments (e.g. Ridgway and Shimmield, 2002). However, the choice of applied techniques strongly 5 depends on the type of pollutants. The disposed mine tailings comprise large quantities of processed geological 6 material, usually enriched in concentrations of extracted metal(s). Therefore, the basic method used include analyzing 7 multi-element geochemistry (e.g. Edinger et al., 2007; Elberling et al., 2003; Larsen et al., 2001; Little et al., 2015; 8 Odhiambo et al., 1996; Perner et al., 2010), often supplemented by analyses of organic carbon content and grain-size 9 distribution, both being closely interrelated with most of the investigated elements/metals (Elberling et al., 2003; 10 Edinger et al., 2007; Little et al., 2015). They are occasionally supported by radionuclide activity measurements to 11 provide an independent time scale (e.g. Elberling et al., 2003; Odhiambo et al., 1996; Little et al., 2015; Perner et al., 12 2010). In a perspective of the processes governing the spreading and potential impacts of the discharged mine tailings 13 on the marine environment it seems equally important to track the distribution of the tailing particles themselves, and 14 not only the tailing-related contaminants. Grain-size analysis may provide additional evidence to understand material 15 dispersion pattern, as exemplified by Okada et al. (2009) in their research on the spatial distribution of dredged material 16 disposed in UK coastal waters. 17

The main aim of this study is to assess the state of the sedimentological environment and the potential impacts of submarine tailings disposal from copper (Cu) mine activities on the fjord Repparfjorden (northern Norway), nearly 40 years after its cessation. The fjord has two basins, the inner and outer basin separated by a sill. Mine tailings were discharged into the inner part of the fjord during copper mining operations in the 1970s in an amount estimated at ~1 million tons (Kvassnes and Iversen 2013).

Previous investigations of Repparfjorden are sparse and only reported in grey literature and in consultancy reports 24 25 (Christensen et al., 2011; Dahl-Hansen and Velvin, 2008). These authors mainly examine surface sediment samples, characterizing primarily the present state of the Repparfjorden environment. They document elevated concentrations of 26 27 heavy metals in surface sediments in the vicinity of the discharge point, while finding no significant differences between the inner and outer fjord in benthic organisms or in benthic biodiversity. In addition, one ²¹⁰Pb-dated core 28 29 collected from the central part of the fjord (Fig. 1) was investigated for heavy metal concentrations revealing that only 30 pollution of a negligible level occurred in surface sediments (Christensen et al., 2011). The characteristics of the tailings 31 discharged during the 1970's were partly a focus of Pedersen et al. (2016) in terms of metal availability and its potential 32 mobilization. In their study, a 10-cm-thick sediment sample taken 5 cm below the seafloor from the inner fjord was 33 investigated, assuming to be representative of the mine tailings. Chemical analyses revealed a relatively high Cu 34 concentration that was associated with more available fractions.

Here we examine the sedimentological and geochemical records obtained from seventeen short sediment cores retrieved from the entire fjord. We give particular attention to the reconstruction of sedimentation based on grain-size characteristics and sediment accumulation rates, as well as potential sediment transport paths with prime focus on lateral and vertical spreading of mine-tailing related contaminants (metals) and particles. In light of the obtained results, we also address questions of potential long-term impacts of environmental pollution in the fjord.

2. Study area

Repparfjorden, located in Finnmark County (northern Norway) is an approximately 13 km long, up to 4 km wide and c. 37 km² large fjord (Fig. 1). Water exchange with the open ocean occurs through Kvalsundet and Sammelsundet. The fjord comprises two main basins, a smaller basin with maximum water depth of c. 65 m in the inner fjord and a larger basin with up to c. 120 m depth of the outer fjord. They are separated by an E-W orientated sill of approximately 50 m depth. An additional sill, interpreted to be part of an end moraine (Marthinussen, 1960) crosses the fjord mouth. Repparfjorden is a temperate non-glaciated river-influenced fjord, with the main river Repparfjordelva at the fjord head. This river drains an area of 1019 km². A delta and a tidal flat occur at the transition from the river to the fjord.

51 Copper deposits of the Ulveryggen and Nussir ore formations occur south of Repparfjorden. They are associated with 52 the Repparfjord Tectonic Window within the Caledonides. The first mining activities of Repparfjorden ore deposits 53 commenced in the beginning of the 20th century, however, large-scale operations started in the 1970s. Between 1972 54 and 1978 (1979) approximately 3 Mt of ore were mined from open pits in the Ulveryggen formation (Sandstad et al., 55 2012) and approximately 1 million tons of tailings were subsequently discharged at the inner ford bottom (Kvassnes 56 and Iversen, 2013). The exact composition of the tailings before disposal (particle size, geochemical characterization 57 and chemicals used) along with the location of the discharge outlet are uncertain and the information provided below 58 are based on disparate sources, namely brochures issued by the mining company in the 1970s and community 59 interviews (Svendsen J., pers. comm.). The mine tailings were discharged about 1.5 km out from the processing plant 60 through a pipeline at a depth of about 60 m (for location see Fig. 1C). The pipeline, suspended 20 m above the seafloor, 61 had openings at its outlet over a length of 600 m spaced at intervals of 100 m. About 80% of the ore was milled to the 62

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size of 74 µm, subjected to the flotation process and thereafter mixed with flocculent chemical and disposed at the fjord bottom. The flocculent (likely Separan NP10) was used to prevent mobilization and facilitate rapid settling of the tailing particles. According to recently conducted chemical characterization of tailings from the Ulveryggen deposits, that were produced using similar methods as in the 1970s, the Cu concentrations varies between 707 and 1090 mg/kg depending on the grain-size fraction of analyzed subsamples (Kleiv, 2011).

3. Material and methods 3.1. Sampling procedure

The material for this study was collected during two cruises in April 2013 and June 2015 with R/V Helmer Hanssen of UiT The Arctic University of Norway in Tromsø (UiT). Sediment cores, up to 21 cm long, were taken from seventeen stations covering the entire fjord (Fig. 1, Table 1). They were retrieved with a multi corer (MC) or with a box corer (BC), respectively. The MC contains six plastic liners (up to 80 cm long; 11 cm outer diameter), whereas BC consists of a metal box with dimensions of 50 cm \times 50 cm \times 50 cm into which plastic liners were pushed to sub-sample individual sediment cores. The surfaces of all of the cores analyzed in this study were undisturbed. Whenever possible a duplicate of each core was retrieved to ensure enough amount of (dry) material for the purpose of this study (~40 g) as well as for the foraminifera analysis (preferable >100 g) (Skirbekk et al., in prep.) and detailed chemical analyses (~30 g) (Pedersen et al., sub.). The seventeen cores together with one of their duplicates were sliced into samples of 1-cm thickness and frozen immediately after retrieval. All samples were freeze-dried prior to the analyses (for an overview of analyses see Table 1). Unless otherwise stated, each core together with its duplicate will be referred by a short name according to the following pattern, e.g. HH13-001-MC-MF-D, hereafter named 001, and IG15-1-1039-BCA, hereafter named 1039, etc.

3.2. Sedimentological properties

Grain-size analyses were performed on all samples at the Department of Geosciences at UiT (Table 1). The sub-samples (2 g) pre-treatment followed the procedures described by Dijkstra et al. (2016). The grain-size measurements were carried out with a Laser Diffraction Particle Size Analyzer (Beckman Coulter LS 13 320) with Polarisation Intensity Differential Scattering (PIDS) analyzing the range from 0.017 to 2000 µm. The measurements were performed in three runs of 60 seconds and the average was calculated subsequently. Sediments classification is according to Folk's (1954) scheme, whereas grain-size statistical parameters were calculated with the GRADISTAT software (Blott and Pye, 2001) applying the geometric method of moments.

3.3. Sediment geochemistry

Heavy metal concentrations were determined for all pulverized sediment sub-samples (2 g) of fractions <2 mm (Table 1). Measurements of As, Ba, Cd, Co, Cr, Cu, Ni, Pb, Ti, V and Zn concentrations in samples from 14 cores were conducted at Unilab Analyse AS via the sub-contracted ALS Laboratory Group Norway AS, using a inductively coupled plasma atomic emission spectrometer (ICP-AES) or inductively coupled plasma sector field spectrometer (ICP-SFMS) following nitric acid digestion. Mercury concentrations were determined using atomic fluorescence spectrometer (AFS). In both cases, standard procedures following Norwegian Standard 4770 and 4768 were applied (Norwegian Standard 1994, 1989, respectively). Metal concentrations of As, Ba, Cd, Co, Cr, Cu, Ni, Pb, V and Zn in samples from 3 cores were analysed at the Technical University of Denmark (DTU), also based on acid digestion and using inductively coupled plasma - optical emission spectrometry (ICP-OES) following Norwegian Standard 4770 (Norwegian Standard 1994).

Total organic carbon (TOC) was measured in bulk sediments for all sub-samples (2 g) at UiT and at DTU (Table 1). The TOC content was calculated accordingly TOC = TC – TIC, where TC is total carbon and TIC is total inorganic carbon content. At both laboratories, TC was measured using a LECO CS-200 instrument. At UiT, the content of TIC was measured using the LECO CS-200 instrument after removing calcium carbonates from the samples with ~20% HCl. At DTU, TIC measurements were carried out in a Scheibler apparatus after dissolving calcium carbonates with ~10% HCl.

3.4. Sediment accumulation rates

The assessment of modern sediment accumulation rates (SARs) was based on the interpretation of ²¹⁰Pb and ¹³⁷Cs activity profiles of eight cores retrieved from both the inner and outer fjord (Fig. 1, Table 1). The activities were measured by gamma spectroscopy using a high-purity germanium detector (Canberra GX2520) at the Institute of Geology, Adam Mickiewicz University in Poznań (Poland) following procedures described by Szczuciński et al. (2013). Each freeze-dried sub-sample (up to 30 g) was homogenized and packed in a plastic sealed container, thereafter stored for several weeks. To enable the calculations of SARs excess ²¹⁰Pb activities (²¹⁰Pb_{ex}) were determined by subtracting the ²¹⁰Pb supported activity (²¹⁰Pb_{supp}) (average of ²¹⁴Pb, ²¹⁴Bi and ²²⁶Ra) from the total ²¹⁰Pb activity (²¹⁰Pb_{tot}).

To calculate SARs the Constant Initial Concentration model was used (for review see e.g. Carroll and Lerche, 2003) assuming that the ²¹⁰Pb_{ex} concentration in surface sediments is constant over time and the ²¹⁰Pb_{ex} flux to the surface of the sediments together with the accumulation rate are proportionally variable. The assumption of a closed system in this model imposes that the ²¹⁰Pb_{ex} activity profile shall decrease exponentially with depth. However, non-steady sedimentation caused by mine tailing discharge during the 1970s and sediment surface mixing are probably the main reason for non-exponential down-core decreases of ²¹⁰Pb_{ex} activity with depth (for details see section 4.3). In consequence, ²¹⁰Pb_{ex} activity profiles were divided into intervals representing different accumulation rates and then the average SAR for each interval was calculated, following the equation:

 $SAR = \lambda \times z \times (\ln A_0 / A_z)^{-1}$

where λ is ²¹⁰Pb disintegration constant (=0.03118 yr⁻¹), z is a depth in a core (cm) (in this case thickness of the interval), A₀ is the ²¹⁰Pb_{ex} concentration at the surface (top layer of the interval), and A_z is the ²¹⁰Pb_{ex} concentration at depth z (bottom layer of the interval). To validate our ²¹⁰Pb-derived assessment of SARs we used anthropologically induced ¹³⁷Cs as a time marker with its first occurrence during the early 1950s and maximum activity around 1961 (e.g. Robbins and Edgington, 1975). Due to limitations in the use of this dating method (e.g. non-steady sedimentation and sediment mixing) calculated SARs should be treated as approximate SARs.

3.5. Background heavy metal concentrations

17 In order to assess the degree of heavy metal pollution for Reppartforden sediments containing discharged mine tailings, 18 it is important to first distinguish between natural (background) and anthropogenic metal loads. The natural variation in 19 heavy metals is mainly attributed to variations in sediment grain-size, hence also their mineralogical composition. In 20 general, heavy metals have an affinity for finer sediment fractions (e.g. Barbanti and Bothner, 1993; Brook and Moore, 21 1988; Zonta et al., 1994). Accordingly, the simplest and highly recommended approach to assess the natural local 22 background levels is to determine heavy metal concentrations in texturally equivalent subsurface sediment core samples 23 obtained from a depth below any possible contamination or biological mixing (Loring and Rantala, 1992). To meet the 24 "pristine" criterion in this study a twofold approach was applied. Firstly, the sediments were confirmed as being 25 deposited before tailings disposal. We identified core sections deposited before the year 1970 using our derived SARs. 26 These sections were inspected for heavy metal concentrations to identify the lowest concentrations, considered to reflect 27 the least impacted levels. Using this procedure, four cores were chosen: 002, 003, 005 and 1075. In order to fulfil the 28 second condition of textural equivalence three background metal levels are proposed. They were determined by 29 averaging the three lowest concentrations of a specific heavy metal found in the four chosen cores in an equivalent 30 textural sediment group, namely of comparable mud content (Table 2). 31

To validate our estimations, we compared obtained background levels with available geochemical data from Repparfjorden (Christensen et al., 2011) (Fig. 1). According to Christensen et al. (2011) the lowest measured concentrations for Cd, Cu, Pb and Zn were found in the deepest sample (14-15 cm) consisting of 73% mud and ²¹⁰Pbdated to the year 1939. The concentrations (Table 2) are of comparable values with our background level 1, estimated for samples composed of 66% mud on average.

38 The major portion of sediments in temperate fjords, like Repparfjorden, derives from fluvial sources (Syvitski et al., 39 1987). Therefore, in the assessment of natural heavy metal concentrations it is also crucial to consider the natural 40 chemical composition of sediments from the Repparfjordelva catchment. For this purpose, we used the chemical 41 composition of overbank sediments (floodplain sediments) (Ottesen et al., 2000), since they are regarded the most 42 representative for geochemistry of the drainage area (Ottesen et al., 1989). As presented in Table 2, the metal 43 concentrations in overbank sediments (Ottesen et al., 2000) are one order of magnitude higher in some cases compared 44 to the samples presented in this study. This is likely due to the usage of different sediment fractions. The metal 45 concentrations in overbank sediments were analyzed in the mud fraction ($<63 \mu m$), whereas in our study concentrations 46 were measured in a gravel-free fraction (< 2 mm). However, if we recalculate proportionally the percentage content of 47 mud fraction to account for grain-size variations, our estimates are more comparable to the concentrations of overbank 48 sediments. 49

3.6. Geoaccumulation index

The degree of heavy metal contamination of the sediments was assessed using the geoaccumulation index (I_{geo}) proposed by Müller (1969) and defined as:

 $I_{geo} = \log_2 \left(C_n / 1.5 B_n \right)$

where C_n is a measured concentration of a given metal in sediment layer X, B_n represents background concentration of a given metal either found in the literature or measured in texturally equivalent uncontaminated sediments. To meet the criterion of textural equivalence, the established background levels (see section 3.5 and Table 3) are a function of the mud content in the sediment sample for which the index was calculated. A factor of 1.5 is introduced to account for possible variations in background values. The level of contamination is subsequently based on increasing values of

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calculated I_{geo} and presented as seven descriptive classes with the highest class representing 100-fold enrichment above background values (Table 3).

3.7. Statistical analysis

Pearson correlation was calculated for standardized data using the Past 3.06 software. The transformation applied for samples of each core separately brings all values to compatible units of a distribution with a mean of 0 and a standard deviation of 1. The statistical significance of the correlation coefficient (r) is expressed by the two-tailed probability (p) where p <0.01 results are considered to be of high statistical significance, 0.01 0.05 of intermediate significance and p >0.05 of low significance. Cluster analysis was performed with the Statistica 10 software to reveal similarities in sedimentological and geochemical compositions among the sediment samples. We used the Euclidean distance and the Ward method on the raw data set.

4. Results

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4.1. Sedimentological properties

15 Fjord sediments are generally fine-grained and moderately to very poorly sorted (Fig. 2 and Supp. Fig. 1 and Supp. 16 Table 1). The sand content in all analyzed samples varies between 1.8 and 79.8%, the silt fraction ranges from 18.6 to 17 85.4%, and the clay content rarely exceeds 10% (Table 4). Although the contents of sand and silt fractions vary the 18 most, the majority of the samples, 86.4% on average, is mostly composed of medium silt to fine sand fractions (8-250 19 μm) (Supp. Table 1). Based on the Folk's (1954) scheme the samples are classified into three textural groups: silty sand 20 (n=96), sandy silt (n=165) and silt (n=11) (Fig. 3). In general, coarser sediments characterize samples from the outer 21 fjord (Fig. 3). Silty sand sediments dominate mostly at the SW part of the outer fjord and comprise all samples of cores 22 007, 010, 011 and 013 as well as the majority of samples from the bottom part of core 005 and the top part of core 1065. 23 They are also found in cores from the NE part of the outer fjord and constitute 50% of samples in core 008, 17% in core 24 1075 and 13% in core 1087. Moreover, single samples are found in cores 002 and 1039. Sandy silt type sediments 25 dominate in the NE outer fjord as well as in the inner fjord. In the NE outer fjord they constitute all samples of cores 26 003, 004, 009 as well as 87% of samples in core 1087, 83% in core 1075 and 50% in core 008. In the inner fjord, sandy 27 silt sediments comprise all samples of core 1089, 95% of samples in core 002, 93% in core 1039, 70% in core 1079 and 28 60% in core 001. The least common silt type dominates in the inner fjord, specifically in the bottom part of core 001 29 (40% of samples) and at 6-13 cm depth in core 1079 (30% of samples). A single sample of silt type is also found at the 30 bottom of core 1065 from the SW outer fjord. The frequency curves of sediment samples plotted as depth profiles for 31 each core show nearly uniform trends with dominating unimodal grain size distribution (Fig. 4 and Supp. Fig. 2). In 32 cores 001, 002, 1039, 1079 and 1089, retrieved from the inner fjord, an interval of bimodal distribution is found with 33 one mode within the very coarse and one mode in the coarse silt fraction (31-63 µm and 16-31 µm, respectively). This 34 interval is typically a few centimeters thick, mostly found at core depths between 3 and 14 cm, and it is more fine-35 grained compared to the over- and under-lying sediments. A similar pattern of increased mud content combined with 36 non-unimodal distribution was also found in a few samples from the lowest part of cores 1065 and 1087 from the outer 37 fjord. However, those sediments are of tri- to polymodal distribution. 38

4.2. Sediment geochemistry

The TOC contents vary between 0.03 to 1.9 wt% (Table 4). The highest average contents, i.e. >1 wt% occur in cores 001, 002, 003, 005, 009 and 011 (Fig. 2 and Supp. Fig. 1). In general, vertical changes of TOC content in most of the cores are relatively small. They typically increase slightly towards the core tops. The lowest TOC contents occur in an up to 10 cm thick interval terminating about 3 cm below the surface of the following cores from the inner fjord: 001, 002, 1039 and 1079.

46 The highest average concentrations (tens to hundreds of mg kg⁻¹) in all analyzed samples have Ni, V, Zn, Cr, Ba, Cu 47 and Ti, whereas average concentrations of Pb, Co, As, Cd and Hg are lower (Table 4). The Hg concentration in 48% of 48 the samples was below its detection limit (0.01 mg kg⁻¹) (Table 4, Fig. 2 and Supp. Fig. 1). Highest average 49 concentrations of heavy metals occur in cores retrieved from the inner part of the fjord: As, Ba, Cd, Cu and Zn in core 50 1079; Pb, Ti and V in core 1089; Co, Cr, Ni in core 001; Hg in core 1039. Concentrations of Cu are the most variable 51 for all measured heavy metals, in particular in cores from the inner fjord: 002, 1079, 1089, and in one core 003 from the 52 outer fjord, where concentrations increase in an up to 10 cm thick interval 4-8 cm below the seafloor (Fig. 2 and Supp. 53 Fig. 1). Pearson correlation calculated for all samples reveals strong positive correlations (r > 0.7) of high significance 54 levels (p < 0.01) in two distinct groups of metals: Ba-Co-Cr-Cu-Ni and Pb-V-Zn (Table 5). Correlations calculated for 55 56 samples of individual cores show the same pattern in cores 002, 1079 and 1089 from the inner fjord. A pattern of two similar distinct groups: Co-Cr-Cu-Ni and As-Pb-Ti-V-Zn (r > 0.7 and p < 0.01) is observed in cores 001 and 1039 (Supp. 57 Supp. Table 2), also from the inner fjord. In the case of the cores from the outer fjord, correlations between measured 58 metal concentrations are more diverse. However, in general they show positive correlations of r > 0.6 and p < 0.01 for a 59 majority of metals, most often between Ba, Co, Cr, Cu, Hg, Ni, Pb, Ti, V and Zn. As and Cd show predominantly weak 60 61 negative correlations of intermediate to low statistical relevance to the rest of the metals.

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4.3. Geoaccumulation index

According to the classification based on the I_{geo} values after Müller (1969) (Table 3) the majority of highly contaminated sediments (classes >4) is found in the inner part of the fjord, whereas sediments from the outer fjord are mostly uncontaminated to moderately contaminated (classes <1) (Fig. 2 and Suppl. Fig. 1). Moreover, the concentrations of metals like Co, Pb, Ti, V and Zn reveal uncontaminated to moderate contamination (classes <1) in all of the investigated cores. Very few samples indicate moderate to strong contamination (classes 2 to 4) by Cd, Cu and Hg in cores 003, 1065 and 1087 from the outer fjord. In addition to a few moderately contaminated (class 2) samples by Cd, Cu and Hg in core 009 there is also one sample (9-10 cm depth) with moderate to very high contamination (classes 3-5) by all heavy metals excluding Hg. The highest contamination levels in sediments from the inner fjord are caused by Cu with strong to very high contamination (classes >4) detected at approximately 5-13 cm depth in cores 002, 1079 and 1089 as well as in the entire cores 001 and 1039. A similar distribution pattern but of lower contamination degree (classes 2-4) is also characteristic for Ba and Cr. Moreover, moderate to strong contamination (classes 2 and 3) by Hg was found in a majority of the sediment samples from this part of the fjord, whereas contamination of classes 3 and 4 by Cd is only seen in core 1079.

4.4. Sediment accumulation rates

All of the ²¹⁰Pb_{ex} activity profiles generally decrease with sediment depth but with near-uniform and/or decreased activities in surface sediments (\leq 5-cm), representing a surface mixed layer (e.g. Jaeger et al., 1998) (Fig. 5). In addition, uniformly low activities occurred within approximately 7-cm-thick intervals ~6 cm below the tops of core 002 and 1079 from the inner fjord, representing depositional events (e.g. Jaeger et al., 1998). There is also a clear difference in measured activities at the core tops (\leq 5-cm) between cores 001, 002, 003, 1079 and cores 005, 013, 1065, 1075. The ²¹⁰Pb_{ex} activities in the first set of cores are twice as high on average compared to the second set.

Table 6 summarizes the SARs that were calculated for intervals below the surface mixing layer. In cases where no significant changes in the shape of the ²¹⁰Pb_{ex} activity profiles were recognized, i.e. in cores 003, 005, 013, 1065 and 1075, the ²¹⁰Pbex values for the deepest interval were used resulting in the following SARs 1.1, 1.5, 0.7, 0.9 and 0.9 mm yr⁻¹, respectively. In cores form the inner fjord, i.e. 002 and 1079, with evidence of depositional events, SARs were calculated in three intervals to estimate their values before, during and after each event. They are as follows, core 002: 2.1, 2.5 and 1.9 mm yr⁻¹, core 1079: 3.6, 4.3 and 1.2 mm yr⁻¹, respectively. Core 001, also retrieved from the inner fjord, is only 10 cm long so that its ²¹⁰Pbex activity profile is incomplete, thus making the calculations of SARs using radionuclide methods less accurate. However, as an analogue to cores 002 and 1079 the lowermost sediments below 6 cm depth are interpreted as the uppermost parts of an event-deposition interval, allowing the estimation of the after-event-deposition SAR. This calculated SAR of 1.5 mm yr⁻¹ is in good agreement with equivalent SARs estimated in cores 002 and 1079.

Similar to ²¹⁰Pb_{ex} the ¹³⁷Cs activities are generally higher in cores 001, 002, 003, 1079 compared to core 005, 013, 1065, 1075 (Fig. 5). Clear peaks in ¹³⁷Cs activities that could be related to the intensification of nuclear bomb testing at the beginning of 1960 or the Chernobyl disaster in 1986 are absent. Most profiles are blurred in shape, although some elevated activities in a peak-like form are visible in profiles of cores 001, 002, 003 and 1079. The observed peak-like features are assumed to be artefacts caused by increased SARs and surface mixing of the sediments, hence diluting ¹³⁷Cs activities. ¹³⁷Cs-derived SARs were calculated based on maximum penetration depth of ¹³⁷Cs excluding the depth of surface mixing layer recognized in ²¹⁰Pbex activity profiles. Although, some of the cores are characterized by deep penetration of low ¹³⁷Cs activities, possibly related to deep mixing/bioturbation, estimated SARs by both ²¹⁰Pb and ¹³⁷Cs methods are generally in good agreement (Table 6).

5. Discussion

5.1. The state of the sedimentological environment and heavy metal contamination levels

The inner and outer parts of Repparfjorden are characterized by two distinct sediment dispersal patterns. The inner fjord acts as a major depo-centre of sediments. Pre- and post-event deposition SARs in the inner basin are on average one order of magnitude higher than in the outer fjord (Table 6) and sediments are generally finer (Fig. 3). A doubling of 210 Pb_{ex} and 137 Cs activities (Fig. 5) in sediment cores from the inner to outer fjord indicates an additional large source of radionuclides from sediments entering from the river, causing a so-called focusing effect (Appleby, 1998). A similar pattern of increased radionuclide activities as well as dominance of finer sediments is also observed in core 003 from the outer fjord, in the vicinity of the sill. This indicates the strongest influence of riverine discharge among the investigated sediment cores of the outer fjord. Calculated SARs in the outer part of Repparfjorden (Table 6) are generally comparable to SARs reported for four marine sites in Finnmark County from Sørøya, Snefjord,

Porsangerfjorden (0.45-0.95 mm yr⁻¹) and Magerøya (0.64 up to 2.8 mm yr⁻¹) (Larsen et al., 2010). In light of these results, the average SAR of 3.8 mm yr⁻¹ calculated for a sediment core from central Repparfjorden (Christensen et al., 2011) (Fig. 1) may be overestimated.

The generally coarser granulometric composition in the outer fjord sediments (Fig. 2 and 3, Suppl. Fig. 1), combined with lower SARs (Table 6), implies reduced sediment supply and/or a more intensive bottom current flow in this area leading to non-deposition or winnowing of fine-grained material. The dominance of finer sediments - sandy silt type at the northeastern part and coarser - silty sand at the southwestern part of the fjord would suggest relatively stronger bottom currents at the southern side of the outer fjord. Christensen et al. (2011), conducted in-situ measurements of bottom-current intensity for two short periods at nine stations along the southern and western shores of the fjord. Stronger bottom currents were observed during the summer period with a predominately inflow direction. The observed asymmetrical pattern in granulometric composition of the outer fjord sediments probably also reflect the influence of the Coriolis Effect, leading to the counter-clockwise circulation (for details see e.g. Cottier et al., 2010). Suspended/turbid sediments derived from Repparfjordelva are deflected to the right, looking towards the fjord mouth, resulting in the observed transport/deposition of the sediments along the eastern and northern shores of the fjord. In summary, it is suggested that the major portion of net outflow and major sediment transport at the bottom of the outer fjord occurs along the northeastern side (compare with Plassen and Vorren, 2002). 16

17 Norwegian Pollution Control Authorities classify contaminants in marine sediments for ecotoxicological effects (SFT, 18 2007; Bakke et al., 2010). This classification system was developed on the basis of previous guidelines taking into 19 account the statistical distribution of contaminant levels found in soft sediments along coastal regions in Norway and is 20 in accordance with the risk assessment principles of the European Union (for details see Bakke et al., 2010). Five levels 21 of contamination are distinguished for the priority metals As, Cd, Co, Cr, Hg, Ni, Pb and Zn. Whereas class I is defined 22 as background contaminant level, classes II to V represent different levels of probable toxicity (Table 7). The border 23 between classes II and III is of critical importance for the assessment of sediment quality since it separates non-toxic 24 and toxic contamination levels (Bakke et al., 2010). This classification system was developed for fine-grained 25 sediments (mud fraction) and is regarded as not suitable for sediments containing gravel and coarse sand fractions. We 26 apply this classification in our study because the investigated sediment samples are dominantly composed of mud (clay 27 and silt) fraction (Table 4, Fig. 3) with an average content of 59.7%, whereas the majority of sand fraction is very fine 28 to fine (63-250 µm) (Supp. Table 1). The coarse and very coarse sand fractions (0.5-2 mm) rarely exceed 1% on 29 average and are only found in four cores (002, 004, 1065 and 1087). Moreover, we suggest that the assessment of 30 sediment quality should consider bulk sediments as that is how they are found in the natural environment. 31

32 In our study, the majority of the investigated sediments corresponds with class I representing background levels 33 following the Igeo-based classification indicating uncontaminated to moderately contaminated sediments (Fig. 2 and 34 Suppl. Fig. 1). Moreover, in almost all cases of moderate to strong contamination for As, Cd, Cr, Hg, and Ni those 35 levels are potentially not toxic for organisms according to the Norwegian sediment quality classification system (SFT, 36 2007). One of the exceptions is a sample from 9-10 cm depth of core 009 that represents class IV (bad state) for Cu and 37 Ni. However, Cu causes the highest and most frequent degree of contamination corresponding to classes IV and V, 38 representing levels potentially posing toxic effects after only short-term exposure. This is found in an approximately 13 39 40 cm-thick surface layer of all sediment cores from the inner fjord (Fig. 2 and Suppl. Fig. 1). In addition, in core 003 from 41 the outer fjord a 2 cm-thick sediment layer at ~7 cm core depth is of bad quality (class IV) due to Cu contamination. No 42 similar elevated levels of Cu were found in a core SED kjerne (Christensen et al., 2011) retrieved near core 003 (Fig. 1). 43

5.1.1. Tailings dispersal record

The geochemical and sedimentological records reveal some similar trends for the entire study area. A cluster analysis was carried out to elucidate the lateral and vertical relationships among the studied variables. The set of samples used for the cluster analysis includes all samples for which grain size, heavy metals and TOC concentrations were acquired. Only concentrations of Ba, Co, Cr, Cu, Ni, Pb, V, Zn were chosen due to their specific correlation pattern (see section 4.2).

52 The cluster analysis revealed two main clusters A and B. In relation to the linkage distance the B cluster was also 53 subdivided into second-order sub-clusters: 1, 2 and 3 (Fig. 6). These cluster results along with the assessment of SARs 54 distinguish three types of sediments: those simultaneously deposited with mine tailings and affected most by the 55 disposal, hereafter called mine-tailing sediments; sediments affected by mine tailing disposal, called tailing-affected 56 sediments; and natural sediments, not affected by the disposal.

57 Mine-tailing sediments. The samples interpreted as mine-tailing sediments are equivalent to sub-clusters B1 and B2 58 (n=8 and n=23, respectively). They are found only in cores from the inner ford, the area of intended tailing disposal, at 59 the depths of 5-9 cm below the seafloor in cores 002, 1079 and 1089 as well as in bottom parts of cores 001 and 1039 60 (3-6 cm depth below the surface, respectively). The dominating anthropogenic origin of those sediments is deduced 61

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from their geochemical signature, that is the highest concentrations of Ba, Co, Cr, Cu and Ni, among which Ba and Cu indicate the highest degree of contamination, up to 760 and 1316 mg/kg (340 and 550 mg/kg on average), respectively (Fig. 2, Suppl. Fig. 1). Following the Norwegian sediment quality classification (SFT, 2007) the observed Cu levels 1 correspond to the highest class of contamination indicating sediments of very bad state with concentration levels that 2 may pose severe acute toxic effects. The presence of elevated Ba content following the pattern of increased Cu 3 concentration is most likely related to the mineralogy of the mined ore formation. In the Repparfjord Tectonic Window 4 area barite (BaSO₄) was reported in vein structures (Viola et al., 2008; Vokes, 1956). Moreover, elevated Ba 5 concentrations, 551 mg/kg on average, characterize the newly processed tailings from the Ulveryggen formation (Kleiv, 6 2011). Alternative potential source of Ba is drill cuttings disposed in the old open pits of the Ulveryggen mine since 7 2005 (Ettner and Sanne, 2016). The Ba-based components are often used by petroleum industry at seafloor extraction 8 sites as a flocculent in drilling mud (for details see e.g. Lepland et al., 2000). However, there is no indication of elevated 9 Ba concentrations in the surface fjord sediments deposited during the last decade that could be linked to a potential 10 leakage of the drill cuttings. The occurrence of event-deposition layers with high SARs at almost the same depths in 11 cores 001, 002 and 1079, the layers deposited during the 1970s (Fig. 2, 5 and Table 6) further supports a tailing-origin. 12 Another geochemical feature of those sediments is the lowest TOC content (Fig. 2, Suppl. Fig. 1) indicating diluted flux 13 of the organic material due to higher SARs and/or suppressed biological activity during the time of deposition. The 14 tailing footprint is also clearly visible through the bimodal grain-size distribution (Fig. 2, 4 and Suppl. Fig. 2) 15 characteristic for the majority of the sediments grouped in cluster B2. This distribution suggests two ways of transport 16 and deposition. One of the modes, mostly the 1st mode (very coarse silt; 31-63 µm) is equivalent in size with a mode of 17 natural sediments recovered in the bottommost sections in a few cores from the inner fjord (see 'natural sediments' 18 paragraph and Fig. 4). Secondly, most often the 2nd mode (predominantly medium silt; 8-16 µm) is in the same fraction 19 size as sediments grouped in sub-cluster B1 that are of unimodal distribution. Samples representing this sub-cluster are 20 only found in core 1079 which was taken in central part of the inner fjord from the site most proximal to the probable 21 tailings disposal outlet (Fig. 1). This pattern is accompanied by the highest concentrations of Cu (up to 1316 mg/kg), 22 which are comparable to the Cu concentrations for the newly produced Ulveryggen tailings (Kleiv, 2011). Therefore, 23 those sediments are interpreted to be mostly composed of mine tailings. Additionally, a specific spatial trend of the 24 bimodal distribution of the sediments from sub-cluster B2 is observed. In cores 001 and 002 taken closer to core 1079 25 (most proximal to the past probable pipeline outlet location), the 1st mode is in the finer silt fraction ('tailings mode'), 26 whereas in more distant cores 1039 and 1089, the 1st mode is found in the coarser silt fraction ('natural mode'). This 27 28 trend is interpreted to reflect the spatially decreasing impact of the discharged tailings particles with increasing distance from the probable outlet location in the inner fjord. In core 1089, the geochemical and sedimentological trends have a 29 30 vertical offset of approximately 2 cm. This is most likely related to the integration of two core duplicates for the analyses performed (Table 1). 31

32 Tailing-affected sediments. The sediments interpreted as tailing-affected represent samples grouped in sub-cluster B3 33 (n=37). These are mostly found in cores from the inner fjord within a few centimeters-thick layer below the mine-tailing 34 sediments, as well as between the top of the mine-tailing sediments and seafloor. They are also found in the outer fjord 35 between 6-8 cm depth in core 003 located close to the sill, as well as in one sample from 9-10 cm depth in core 009 at 36 the northern side of the outer fjord. The geochemical and sedimentological signatures of those sediments indicate that 37 they were accumulated under an influence of disposed tailings. Tailing-affected sediments are characterized by elevated 38 concentrations of Ba, Co, Cr, Cu and Ni (Fig. 2 and Suppl. Fig. 1). The average Cu concentration of 120 mg/kg is 39 equivalent to IV class of sediments (bad state) according to the Norwegian quality classification (SFT, 2007) (Fig. 2 and 40 Suppl. Fig. 1). This level of contamination may pose toxic effects just after short-term exposure. Most of the tailing-41 affected sediments are characterized by prevailing unimodal distribution of 'natural mode' with only a slightly 42 increased content of medium silt dominating in tailings sediments (Fig. 4 and Suppl. Fig. 2). Although one sample of 43 core 009 was grouped in sub-cluster B3, representing tailing-affected sediments, it is not regarded as such. This sample 44 is characterized by high concentrations of all of the measured heavy metals with an exception of Hg (Suppl. Fig. 1). 45 However, the TOC contents and the results of grain-size analysis do not indicate any excursions (Suppl. Fig. 1 and 2). 46 Therefore, this sample is assumed to be an outlier, perhaps accidentally contaminated prior to or during chemical 47 48 analyses.

49 Natural sediments. Sediments regarded as not affected by the tailings discharge, representing natural fjord sediments, 50 are grouped in cluster A (n=193). They constitute the majority of the investigated samples from cores taken in the outer 51 part of the fjord, as well as some sediment samples from lower parts of cores from the inner fjord (cores 002, 1079 and 52 1089). The geochemical signature of those sediments indicates that they are of natural background concentrations levels 53 (Fig. 2, Table 2) and, therefore, reflect local provenance. The observed variations in heavy metal concentrations are 54 mainly attributed to the varying grain-size composition (most likely related to the circulation pattern suggested 55 previously), a dependence that is also a natural feature of sediments (Loring, 1991). The down-core frequency profiles 56 show a dominating uniform unimodal pattern with the 1st mode in very coarse silt (31-63 µm) or very fine sand (63-125 57 µm) fractions (Fig. 4, Supp. Fig. 2). Only the bottom sections of cores 1065 and 1087 contain sediments of tri- to 58 polymodal distributions (Fig. 4 and Suppl. Fig. 2). Those samples are composed mostly of finer sediments and are also 59 slightly enriched in almost all of the measured heavy metals (Fig. 2), reflecting the affinity of metals to the finer 60 fractions. It is assumed that the presence of those sediments represents a disturbance episode, most likely of 61

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anthropogenic origin (e.g. anchoring, fishing) or a record of a gravity flow deposits in case of core 1087 located close to the shore, rather than sediments of different provenance.

5.2. The impact of mine tailings disposal

The impact of discharged mine tailings in Repparfjorden is mainly limited to a relatively small area of the inner fjord, the intended disposal site. The mine-tailing sediments restricted to the inner fjord are found along a ~1.5 km long transect. Sediments classified as tailing-affected are also found in a discrete 2-cm thick layer 6 cm below the top of core 003 from the outer fjord, indicating lateral spreading of tailings away from the discharge point. Core 003 is the most proximal to the sill among all cores from the outer fjord. Neither geochemical nor sedimentological records of the rest of the cores from the outer fjord, including the record of core SED kjerne (Christensen et al., 2011) (for location see Fig. 1), show potential influence of the tailings. Therefore, it is inferred that the spreading is spatially limited to the area about 1 km off the sill and about 2 km out from the probable discharge outlet. According to the recalculated SAR the spreading recorded in core 003 commenced before 1972 (Fig. 2). The ²¹⁰Pb_{ex} activity profile of this core shows 5-cm thick surface mixing layer (Fig. 5) limiting the usage of the radionuclide method for exact SAR calculations, hence also for dating purposes (see section 3.4). Since there are no other documented sources of Cu in the Repparfjorden area (with metal loads similar to level of the tailings) that could potentially have been active before the 1970s, it is deduced that the spreading actually took place during the time of disposal.

The tailings had the biggest influence on the investigated sediments of the inner ford during the time of discharge, causing increased SARs and changes in sediment properties (e.g. bimodality). Furthermore, the geochemical record indicates the highest reported heavy metal concentrations in the same layers, mainly represented by Cu. However, even if the impacts of the tailings are restricted in the lateral dimension, the effects of tailings disposal are still significant, especially in the vertical dimension, i.e. in tailing-affected sediments found below and above mine-tailing sediments. The impact of tailings, visible in the geochemical record, is even found in the surface sediment layers, where Cu concentrations remain at an elevated level, representing bad sediment quality according to the Norwegian quality classification (SFT, 2007). On the other hand, the sedimentological properties of those sediments are more similar to the natural pre-disposal sediments. The observed pattern of the tailings impact decreasing gradually towards the top sections of inner fjord sediment cores indicates physical and/or biological reworking of the sediments after and/or during deposition. After disposal, resuspended mine-tailing sediments (due to natural processes) may release heavy metals to the water column either in the form of suspended particulate matter or in dissolved form (Durán et al., 2012; Xu et al., 2015). With the continuous transport and deposition of natural sediments from Repparfjordelva the released metals may afterwards be mixed (and re-precipitated) to sediments and finally deposited. The combination of a stronger geochemical and weaker sedimentological signal may also indicate remobilization (desorption) and dispersion of Cu within the water phase. Some of the metals, like Cu, have been reported to diffuse out and be transported within the sediment column through pore water under specific conditions (Blasco et al., 2000; Cheevaporn et al., 1995). Moreover, the study of Pedersen et al. (2016) on Cu availability in a 10-cm-thick sediment sample from the inner fjord indicate that about 80% of this metal is bound to more available fractions that are exchangeable, oxidisable and reducible. Consequently, the diagenetic processes like metal diffusion cannot be excluded. However, more detailed chemical studies are needed to elucidate the observed pattern.

From a toxicity perspective the ecological communities of the inner fjord have been continuously exposed to Cu concentrations that may pose toxic effects after short-term exposure for nearly 40 years. This level of exposure is potentially of high significance for benthic communities, especially for infaunal organisms. Previous studies on macrofaunal benthic communities in Repparfjorden are limited to surface samples only (Christensen et al., 2011; Dahl-Hansen and Velvin, 2008). In these earlier studies, benthic diversity did not indicate significant differences between the inner and outer fjord, despite, Cu concentrations in surface sediments representing bad sediment quality. There is a need for further studies in a broader scope that would include micro- and infaunal benthic communities, as well as detailed chemical investigations on the binding pattern of metals, their bioavailability and their changes with time and depth.

5.3. Potential long-term effects of tailings disposal

Lateral and vertical spreading of discharged tailings is mainly controlled by two factors: morphology of the fjord bottom and sediment accumulation. The morphological factor has played a crucial role in limiting the lateral redistribution of tailings. Due to the presence of the sill, the inner fiord environment constitutes a semi-enclosed basin, hence also a sink for the mine tailings deposited over time. Although, the bottom current strength in the outer fjord seems to be relatively high, potentially facilitating the spreading of tailings, the sill acts as a natural barrier. We identified only one discrete layer influenced by tailings in the outer fjord. On the contrary, the second factor - sediment accumulation has played a principal role in shaping the vertical distribution of tailings in Repparfjorden. The inner fjord basin constitutes a major depo-centre for (natural) sediments transported by the Repparfjordelva to the head of the fjord, resulting in SARs up to 2-3 times higher than in the outer fjord. This relatively high SAR downgraded the potential

impact of the disposal operations simply by diluting the tailing-related pollutants in the natural sediments. The average post-disposal SAR in the inner fjord is of 1.5 mm yr⁻¹, meaning that after the cessation of the disposal, during the last 37 years (time between the cessation and average timing of core retrieval), the layer of 5.6 cm-thick sediments on average was deposited. During this time, the Cu concentrations in the core taken close to the probable pipeline outlet (core 1079) decrease from 606 mg/kg (the topmost sample of mine-tailing sediments) to 84 mg/kg (the surface tailing-affected sediments). Taking into consideration the average concentration for the entire inner fjord, the Cu concentrations decrease accordingly from 308 to 98 mg/kg.

As postulated by Skei (2013), the prospective submarine disposal sites "should ideally be deep (>100 m), flat or slightly sloping bottom and surrounded by sills to make an enclosed basin where sedimentation is prominent". Those features are mostly met in Repparfjorden. The present study, conducted nearly 40 years after the cessation of tailings disposal, proves that the distribution of tailings is spatially restricted to the intended disposal area. However, the natural characteristics of fjords do not always ensure limited spreading of the discharged mine tailings. Among the most well-studied fjord disposal sites are two sites from British Columbia (Canada): the Portland Canal-Observatory Inlet system where molybdenum-lead-zinc tailings from Kitsault Mine were disposed (e.g. Odhiambo et al., 1996) and Rupert, Holberg and Quatsino Sund system with discharged copper tailings from Island Copper Mine (e.g. Poling et al., 2002); as well as the Uummannaq Fjord complex, West Greenland, where lead-zinc tailings from the Black Angel Mine were disposed (e.g. Perner et al., 2010). A direct comparison of these fjord tailings disposal sites with the present work is not possible, due to different fjords and tailings characteristics, as well as the application of different methods. Nonetheless, in the West Greenland fjord, the tailing-related contaminants were detected 12 km from the mine, with environmental consequences found beyond the intended disposal areas (e.g. Søndergaard et al., 2011).

Looking at a present-to-future scenario, any significant change in the controlling factors has a potential to re-shape the observed impact of tailings in Repparfjorden. Although it is not feasible to speculate about the timing and magnitude of such changes, processes that may influence the fjord bathymetry and water-mass properties, as well as changes in the amount of sediments transported by Repparfjordelva are potentially important in a long-term perspective.

The prospective future spreading of the tailing-contaminated sediments would need to involve a strong enough bottom current to trigger resuspension. Entrainment of contaminated sediments into the water column would need to occur along with a change in the inner-outer fjord water exchange pattern to enable the transport of tailings over the sill and into the outer fjord. The lack of elevated Cu concentrations and finer-grained sediments typical for mine-tailings sediments in the surface layers of the outer fjord indicate that such processes have not taken place since the cessation of tailings disposal. However, to rule out this possibility, there is a need for i) a monitoring survey focused on the present-day Repparfjorden hydrography, as well as ii) a study of the paleo-record (millennial-timescale), to predict possible patterns and rates of exchange between the inner and outer fjord.

The last of the mentioned changes that might influence the future effects of the disposed tailings is a change in the amount of sediments accumulated in the inner part of the fjord. The average post-disposal SAR resulted in an accumulation of about 5.6 cm-thick sediment layer, within which the average Cu concentrations dropped by \sim 70% to 98 mg/kg. If the SAR was 0.5 mm yr⁻¹ lower, similar to the average outer fjord rate, based on linear interpolation between the recorded average Cu concentrations found at different types of sediments in the inner fjord, then the Cu level of 98 mg/kg would be reached in 2034 resulting in an additional ~2-cm thick layer of sediments. On the contrary, if the SAR was higher, similar to the pre-disposal rate, then those Cu levels would have been reached earlier. Although this calculation is a simplification of the natural fjord conditions, even a seemingly small change in SAR of 0.5 mm yr⁻¹ has a large potential to influence the environmental impacts of the disposed tailings on a human-timescale.

6. Conclusions

We assessed the state of the sedimentary environment and the effects of submarine Cu mine tailings disposal in the 1970s in Repparfjorden, northern Norway, based on sedimentological and geochemical analyses of seventeen short sediment cores. We found that:

- The inner and outer parts of the fjord represent two distinct environmental settings. The inner fjord confined by
 a sill is a depo-centre for contaminated mine tailings. The grain-size distribution pattern of sediments in the
 outer fjord suggest counter-clockwise circulation of bottom currents.
- The mine-tailing sediments were found only in cores of the inner fjord along a ~1.5 km long transect. They constitute an up to 9-cm thick layer, 3-9 cm below the core tops. It is composed of mostly silty sediments of bimodal distribution characterized by the highest observed Cu concentrations (up to 1316 mg/kg and 550 mg/kg on average) and low TOC contents. The strongest influence of the mine tailings occurred in the central part of the inner fjord, close to the location of the probable tailing outlet.

- The distribution pattern of tailing-affected sediments implies that dispersion of tailing-related metals and particles is restricted to a relatively small area of the inner fjord to the immediate surrounding sediments and to a discrete layer in one core from the outer fjord most proximal to the sill. These sediments exhibit elevated Cu concentrations (120 mg/kg on average). Cu concentrations in sediment cores from the inner fjord gradually decrease towards the top core sections, implying physical and/or biological reworking of the sediments after and/or during deposition. Moreover, the lack of a clear tailings signal in the sedimentological record indicates that dispersion of Cu in the water phase is likely.
- The presence of tailing-affected sediments in layers up to the sediment-water interface indicate that the ecological communities of the inner fjord have been exposed to concentrations of Cu for nearly 40 years.
- The dispersal of tailings is mainly controlled by two factors: fjord morphology and sedimentation rate. Although, the strength of the bottom currents in the outer fjord seems to be relatively high, potentially facilitating the spreading of the tailings, the sill is acting as an effective natural barrier for the discharged tailings. The relatively high post-disposal SAR in the inner fjord downgraded the potential impact of the tailings disposal by diluting the tailing-related pollutants in the natural sediments.
 - The pattern of bimodal grain-size distribution along with increased content of finer tailing-related fraction in sediments of unimodal distribution facilitated the assessment of tailings discharge impact and allowed to track the spreading of the tailing particles.
 - There is a need for further studies to simultaneously address micro- and infaunal benthic communities and Cu bioavailability in order to provide an insight into the environmental/toxic effects of elevated Cu concentrations observed in sediment cores from the inner fjord. Moreover, a detailed investigation of the present and past hydrography in Repparfjorden is needed to answer the question of the inner-outer fjord water exchange pattern that may in the future contribute to the spreading of tailing-affected sediments.

Acknowledgements

The authors would like to thank the captains and crews of R/V Helmer Hanssen as well as cruise engineers: B.R. Olsen, S. Iversen, and all of the scientific participants of the two cruises for a help in coring and core sampling. T.M. Dahl, I. Hald and K. Monsen kindly assisted with laboratory works. T. Grytå helped with Fig. 1. This study was conducted mainly within the Environmental Waste Management (EWMA) project funded by the Research Council of Norway through NORDSATSING (grant number: 195160), EniNorge AS and the UiT The Arctic University of Norway in Tromsø.

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Table(s)

Table 1. Overview of the coring stations (core-ID, coring gear, sampling date, location, water depth, recovery, number of 1-cm thick samples per core) and overview of analyses performed (grain size; HM: heavy metals concentration; TOC: total organic carbon content; radioisotopes: ²¹⁰Pb and ¹³⁷Cs activities). Analyses of HM and TOC marked with * were conducted at the Technical University of Denmark. The remaining TOC measurements were performed at the Department of Geosciences, UIT The Arctic University of Norway in Tromso, whereas the remaining HM analyses were carried out at Uniba Analyse AS (Tromso, Norway). SED kjerne from Christensen et al. (2011).

	s	S	s															
Analyzed parameters	Grain size, HM, TOC*, radioisotope	Grain size, HM, TOC*, radioisotope	Grain size, HM, TOC*, radioisotope	Grain size, HM, TOC	HM, TOC* Grain size	Grain size, HM, TOC	Grain size, HM, TOC	HM, TOC* Grain size	Grain size, HM, TOC	Grain size HM, TOC*	Grain size, HM, TOC, radioisotopes	Grain size HM, TOC	Grain size, HM* TOC*, radioisotopes	Grain size HM, TOC, radioisotopes	Grain size HM*, TOC*, radioisotopes	Grain size HM*, TOC*	Grain size HM, TOC	Grain size, HM, TOC, radioisotopes
Number of samples	10	21	20	15	20 19	10	8	20 20	11	18 20	13	15 12	19 15	18 16	20 20	15 14	20 20	7
Recovery (cm)	10	21	20	15	20 19	10	8	20 20	11	18 20	13	14.5 12	19 15	18.5 16.5	20 20	14.5 14	20 20	17
Water depth (m)	47	63	86	59	53 53	60	65	83 83	74	114 114	115	44 44	51 51	72 72	61 61	65 65	57 57	88
Approximate core location within the fjord	inner	inner	outer	outer	outer	outer	outer	outer	outer	outer	outer	inner	outer	outer	inner	outer	inner	outer
Longitude (E)	024°17.516'	024°17.737'	024°16.537°	024°14.996'	024°14.041° 024°14.041°	024°12.180'	024°12.916	024°10.108' 024°10.108'	024°09.357	024°08.883' 024°08.883'	024°06.695'	024°17.178° 024°17.178°	024°11.350° 024°11.350°	024°15.079° 024°15.079°	024°17.530° 024°17.530°	024°12.885' 024°12.885'	024°17.999' 024°17.999'	024°15.681'
Latitude (N)	70°27.611°	70°28.383'	70°29.211°	70°29.867'	70°29.388° 70°29.388°	70°30.042°	70°30.427'	70°31.028° 70°31.028°	70°30.552°	70°29.954° 70°29.954°	70°30.886°	70°27.566° 70°27.566°	70°30.075° 70°30.075°	70°29.462' 70°29.462'	70°28.153° 70°28.153°	70°30.561° 70°30.561°	70°28.397' 70°28.397'	70°29.380°
Sampling date	16.04.2013	16.04.2013	16.04.2013	16.04.2013	16.04.2013 16.04.2013	16.04.2013	16.04.2013	16.04.2013 16.04.2013	16.04.2013	16.04.2013 16.04.2013	16.04.2013	23.06.2015 23.06.2015	24.06.2015 24.06.2015	24.06.2015 24.06.2015	24.06.2015 24.06.2015	24.06.2015 24.06.2015	24.06.2015 24.06.2015	2010
Coring gear	multi corer	multi corer	multi corer	multi corer	multi corer	multi corer	multi corer	multi corer	multi corer	multi corer	multi corer	box corer	box corer	multi corer	multi corer	box corer	multi corer	
Core-ID	001	002	003	004	005	007	008	600	010	011	013	1039	1065	1075	1079	1087	1089	
Station No	HH13-001-MC-MF-D	HH13-002-MC-MF-D	HH13-003-MC-MF-D	HH13-004-MC-MF-D	HH13-005-MC-MF-D HH13-005-MC-MF-E	HH13-007-MC-MF-E	HH13-008-MC-MF-D	HH13-009-MC-MF-D HH13-009-MC-MF-E	HH13-010-MC-MF-D	HH13-011-MC-MF-D HH13-011-MC-MF-E	HH13-013-MC-MF	IG15-1-1039-BCA IG15-1-1039-BCB	IG15-1-1065-BCA IG15-1-1065-BCB	IG15-1-1075-MCA IG15-1-1075-MCB	IG15-1-1079-MCA IG15-1-1079-MCB	IG15-1-1087-BCA IG15-1-1087-BCB	IG15-1-1089-MCA IG15-1-1089-MCB	SEDkjerne

Table 2. Heavy metals background values. Levels 1, 2 and 3 are from this study (Hg* background levels equal to its detection limit); SED kjerne from Christensen et al. (2011); overbank sediments from Ottesen et al. (2000).

Background	Mud [%]	As [mg/kg]	Cd [mg/kg]	Co [mg/kg]	Cr [mg/kg]	Cu [mg/kg]	Hg* [mg/kg]	Ni [mg/kg]	Pb [mg/kg]	V [mg/kg]	Zn [mg/kg]	Ba [mg/kg]	Ti [mg/kg]
Level 1 Level 2 Level 3 SED kjerne	66 52 40 73	3.0 2.2 2.3	0.06 0.04 0.06 <0.2	4.2 3.4 3.1	23 20 17 11	10.0 11.2 6.2	0.01 0.01 0.01	13.3 10.1 9.7	4.4 3.6 2.7 3.6	18 15 14	21 16 14 20	32 29 24	700 658 525
Overbank sediments	100	≤6	_	≤25	≤100	≤16	_	≤16	≤16	≤32	≤39	_	_

Table 3. Descriptive classes of the contaminations degree based on the geoaccumulation index (I_{geo}) values (Müller 1969).

I _{geo} value	I _{geo} class	Description
<0	0	uncontaminated
0-1	1	uncontaminated to moderately contaminated
1-2	2	moderately contaminated
2-3	3	moderately to strongly contaminated
3-4	4	strongly contaminated
4-5	5	strongly to extremely contaminated
>5	6	extremely contaminated

Table 4. Statistics for grain-size, total organic carbon content (TOC) and heavy metals (As, Ba, Cd, Co, Cr, Cu, Hg, Ni, Pb, Ti, V and Zn) concentrations for all investigated sediment cores (for locations see Figure 1).

Core-ID	Sand [vol. %]	Silt [vol. %]	Clay [vol.%]	TOC [wt. %]	As [mg/kg]	Cd [mg/kg]	Co [mg/kg]	Cr [mg/kg]	Cu [mg/kg]	Hg [mg/kg]	Ni [mg/kg]	Pb [mg/kg]	V [mg/kg]	Zn [mg/kg]	Ba [mg/kg]	Ti [mg/kg]
001 Min Max Mean Mean	4.8 23.3 12.1 10.9	70.5 83.9 78.8 79.9	6.3 11.3 9.0 9.2	0.26 1.66 1.12	1.9 7.1 5.0	0.01 0.06 0.04 0.04	7.7 9.8 8.5 8.4	68 147 111 109	126.0 310.0 237.3 257.5	0.034 0.047 0.038 0.037	23.9 37.6 31.4 32.0	2.5 9.8 6.5 7.6	16 39 37 31	18 36 33	118 198 155 152	574 1440 978 1110
002 Min Max Mean Median 003 Min Max	11.4 51.6 28.8 27.0 27.0 24.4 36.4 31.0	45.2 78.6 66.8 66.8 59.5 63.6	3.3 6.2 5.7 4.2 4.2	0.55 1.74 1.02 1.02 1.02 0.95 1.39	0.0 0.0 0.0 0.0 0.0 0.0 0 0.0 0 0.0 0 0.0 0 0.0 0 0.0 0 0.0 0 0.0 0 0.0 0 0.0 0 0.0 0 0.0 0 0.0 0 0.0 0 0.0 000000	0.02 0.04 0.04 0.04 0.01 0.01	9.0 9.0 9.0 9.0 9.0 9.0 9.0 9.0 9.0 9.0	17 80 46 42 40 30	13.8 408.0 129.7 68.6 9.1 77.0 30.7	 <0.01 0.035 0.035 0.026 0.028 0.028 0.028 0.072 0.033 	9.1 26.6 17.8 19.0 19.4 119.4	3.4 9.3 6.9 6.9 11.0 11.0 7.6	221 221 227 227 227 227 227 227 227 227	15 35 35 28 28 28 28 28 28	24 247 97 78 31 46	417 1520 789 638 638 610 897
Median 004 Min Max Mean Median	31.8 39.0 45.2 41.6 41.9	63.0 51.2 54.5 54.5	ທ.4 ຕ. ທ.∂.6. ສ.0.0 ໜ	1.36 0.49 0.78 0.58 0.54	3.7 1.7 2.3 2.3	0.06 0.02 0.08 0.05	5.0 3.6 3.6 7.0	29 26 21 20	27.4 8.9 13.1 11.1	0.025 <0.01 0.018 0.015	15.7 9.2 11.1 11.0	7.5 3.6 7.1 4.9 4.6	22 13 15 15	28 17 19 18	44 25 44 28 29 28	797 469 785 593 576
005 Min Max Mean Mean	49.8 67.9 52.5	29.9 45.4 40.5 43.1	2.1 5.0 3.8 4.0	0.86 1.56 1.28 1.29	1.7 3.3 2.5 2.6	0.02 0.09 0.05 0.04	3.0 3.6 3.5	17 26 21 20	6.0 17.4 11.2 9.8	<0.01 0.020 0.016 0.017	9.4 13.6 11.5 11.2	2.5 7.3 6.0	13 19 16	13 32 19	24 39 30	520 685 597 575
007 Min Max Mean Median	54.4 61.7 58.1 58.0	35.6 41.4 38.3 38.3	2.6 4.1 3.6 3.8	0.45 0.70 0.55 0.56	1.4 3.0 2.3 2.3	0.02 0.07 0.04 0.04	3.5 3.5 3.5	16 22 19 18	7.6 13.3 10.7 11.3	<0.01 0.019 0.014 0.013	9.4 11.7 10.5 10.5	3.3 7.1 5.4 6.0	14 18 16	15 23 19	23 32 27 27	415 605 493 481
Min Min Max Mean Median	48.7 53.6 50.5 50.2	42.6 46.8 45.4 45.5	3.8 8.6 7.5 2.5 2.5	0.46 0.66 0.58 0.60	1.4 2.3 1.9 1.8	0.02 0.03 0.02 0.02	3.0 3.3 3.3 3.3	16 20 18 18	10.2 14.7 12.8 13.0	<0.01 0.013 0.012 0.012 0.012	9.9 11.3 10.4 10.2	4.6 5.8 5.5	12 15 14	16 20 18 18	22 30 26	357 453 399 391
yoy Min Max Mean Median	22.1 38.9 28.3 28.5	56.4 70.9 65.5 65.4	4.7 7.9 6.2 5.9	0.82 1.89 1.13	2.0 37.6 5.0 3.0	0.06 2.20 0.25 0.15	3.0 45.0 6.6 4.6	17 259 39 27	6.6 158.0 24.4 15.3	<0.01 0.034 0.022 0.025	10.2 161.0 22.5 14.8	3.1 76.0 10.4 6.8	14 235 33 22	15 269 39 28	22 324 33	369 6340 892 576
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Table 4. Continuation.

Ti [mg/kg]	343 561 454 470	373 904 547 476	324 722 375	390 861 617 572	1111	747 931 869 871	1 1 1 1	1 1 1 1	986 1230 1106 1110
Ba [mg/kg]	21 30 26	18 37 26 25	16 32 23 23	104 183 132 124	26 69 30	32 55 39	46 760 259 115	30 62 38	49 192 89 85
Zn [mg/kg]	15 22 18 18	10 14 13	12 21 16 16	12 23 23 23 23	12 31 16	16 28 20 20	28 55 41	28 50 36	29 36 31
V [mg/kg]	13 17 15 15	12 15 15	11 18 14	11 25 18 20	14 45 21 16	17 25 20	14 27 25	17 35 23 21	24 33 28 28
Pb [mg/kg]	3.4 6.3 5.2 5.5	2.6 6.6 2.2 2.2	3.0 6.7 5.4	1.2 1.4 1.4 8.4	3.4 8.1 4.8 4.7	3.3 7.3 5.0	3.2 11.1 7.9 9.0	3.5 6.7 5.3	6.8 8.3 8.4
Ni [mg/kg]	8.2 11.7 10.2 10.5	9.2 12.3 10.6	8.1 12.7 10.3 10.0	21.9 35.1 29.4 31.3	8.9 31.0 13.4 10.2	10.6 15.6 12.3	20.6 45.7 29.8 25.8	12.4 24.3 16.1 14.2	16.2 28.8 20.2 20.2
Hg [mg/kg]	<pre><0.01 <0.017 0.014 0.015</pre>	<pre><0.01 0.017 0.015 0.015 0.015</pre>	<0.010.0160.0140.014	0.036 0.071 0.052 0.051	1111	<0.01<0.022<0.017<0.018	1 1 1 1		<0.01<0.031<0.023<0.025
Cu [mg/kg]	6.6 16.4 10.9 11.1	4.6 11.4 7.5 6.8	6.4 10.6 7.6 7.6	137.0 326.0 237.6 263.5	8.8 30.9 13.0 9.9	7.9 27.0 14.9 12.5	20.4 1316.1 388.7 175.0	13.9 29.7 19.7 18.8	14.1 326.0 94.0 69.4
Cr [mg/kg]	14 21 17	13 21 17 16	15 23 19 20	59 140 112	18 53 26 21	21 31 25 25	39 147 87 66	23 46 31 28	30 81 47 46
Co [mg/kg]	2.5 3.5 3.1	3.3 3.3 3.3	2.6 3.3 3.3 3.3	6.4 9.3 8.0	2.4 11.8 4.4 2.9	3.4 5.1 3.9 3.9	4.5 9.2 5.8	2.9 8.4 3.6	5.7 8.3 6.5 6.6
Cd [mg/kg]	0.01 0.06 0.03 0.02	0.02 0.09 0.05 0.05	0.02 0.12 0.05 0.04	0.01 0.05 0.02 0.02	0.00 0.22 0.12 0.13	0.02 0.08 0.05 0.05	0.61 0.86 0.72 0.71	0.43 1.09 0.64 0.56	0.02 0.09 0.05 0.05
As [mg/kg]	1.6 3.2 1.8	1.6 4.3 2.7 2.8	1.4 2.9 2.8	0.7 4.7 3.0	0.6 1.9 1.3	2.1 3.3 2.6	3.4 9.1 6.0	3.4 5.7 4.6	3.4 7.1 4.6 1.1
TOC [wt. %]	0.49 0.70 0.60 0.65	1.35 1.73 1.53 1.52	0.54 0.74 0.61 0.59	0.03 1.08 0.52 0.48	0.31 0.61 0.44 0.43	0.48 0.88 0.66 0.63	0.13 1.72 0.90 1.10	0.40 1.19 0.80 0.86	0.76 1.32 0.99 0.93
Clay [vol. %]	3.5 3.5 8.8	2:2 3.0 2.8	1.7 3.9 3.0 2.9	4.5 9.2 7.0 7.5	3.5 18.2 7.2 4.6	2.6 5.2 3.8	4.8 13.0 8.3 7.8	4.1 13.5 6.1 4.9	4.2 5.8 4.2 4.2
Silt [vol. %]	35.7 40.3 37.8 38.4	22.4 30.3 27.0 27.0	18.6 26.7 24.6 25.4	44.3 77.3 69.2 71.2	30.5 75.8 41.9 35.2	43.5 56.0 48.9 48.3	71.3 85.4 78.0 77.4	43.2 66.3 50.4 48.9	66.7 77.0 72.2 72.4
Sand [vol. %]	55.3 61.7 58.7 57.9	65.9 75.4 70.0 70.2	69.8 79.8 72.4 71.8	14.2 51.2 23.7 21.3	5.9 65.6 50.9 59.8	38.8 53.5 47.2 48.0	1.8 23.8 13.6 15.6	20.2 52.7 43.5 46.4	15.4 28.9 22.2 22.2
Core-ID	010 Min Max Mean Median	011 Min Max Mean Median	013 Min Max Mean Median	1039 Min Max Mean Median	1005 Min Max Mean Median	Min Min Max Median	Min Max Mean Median	Min Max Mean Median	1009 Min Max Mean Median

Table 5. Pearson's correlation matrix for heavy metals (As, Ba, Cd, Co, Cr, Cu, Hg, Ni, Pb, Ti, V and Zn) in all investigated samples (for location see Figure 1). Correlation coefficient (r) values are given in the lower triangle of the matrix, where r > 0.7 are highlighted in grey shading. Two-tailed probabilities (p) are given in the upper triangle of the matrix with p > 0.05, regarded as of low statistical significance, highlighted in grey writing.

	As	Ba	Co	Cr	Cu	Hg	Ni	Pb	V	Zn	Ba	Ti
As		0.03	0.00	0.00	0.11	0.91	0.11	0.02	0.00	0.00	0.00	0.00
Ba	0.13		0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Cd	0.19	-0.32		0.01	0.00	0.00	0.00	0.00	0.00	0.96	0.25	0.01
Co	0.19	0.83	-0.16		0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Cr	0.10	0.87	-0.32	0.89		0.00	0.00	0.00	0.00	0.00	0.00	0.00
Cu	0.01	0.85	-0.38	0.79	0.83		0.00	0.00	0.00	0.00	0.00	0.00
Hg	-0.13	0.26	-0.32	0.25	0.28	0.33		0.00	0.00	0.08	0.00	0.00
Ni	0.14	0.84	-0.21	0.94	0.92	0.82	0.26		0.00	0.00	0.00	0.00
Pb	0.23	0.45	-0.30	0.42	0.42	0.51	0.34	0.42		0.00	0.00	0.00
Ti	0.23	0.40	0.00	0.43	0.40	0.20	0.15	0.36	0.46		0.00	0.00
V	0.42	0.56	-0.07	0.64	0.58	0.46	0.23	0.59	0.74	0.74		0.00
Zn	0.26	0.52	-0.16	0.55	0.53	0.50	0.27	0.57	0.83	0.57	0.84	

Table 6. Sediment accumulation rates (SARs) estimated for eight cores (for location see Figure 1). For details see section 3.4 and 4.3.

Core-ID	Intervals used for ²¹⁰ Pb-derived SARs	Sediment accumulation rate [mm yr ⁻¹]					
	estimation [cm depth]	²¹⁰ Pb (min – max)	¹³⁷ Cs				
001	2-5	~1.5 (1.3 – 1.6)	>1.1				
002	2 – 4 3 – 12 12 – 17	1.9 (0.6 – 2.0) 2.5 (2.1 – 2.6) 2.1 (1.4 – 2.6)	>2.9				
003	5-12	1.1 (0.8 – 1.2)	1.4				
005	5-11	1.5 (1.1 – 1.8)	1.3				
013	5-9	0.7(0.4 - 0.8)	1.2				
1065	2-6	0.9 (0.7 – 1.1)	1.5				
1075	2 – 8	0.9 (0.6 – 1.1)	0.8				
1079	2-5 4-16 15-20	1.2 (1.2 – 1.3) 4.3 (3.7 – 4.7) 3.6 (2.7 – 4.1)	>2.7				

Table 7. The Norwegian environmental quality classification of metal contaminants in marine sediments (Bakke et al., 2010 after SFT, 2007).

	I Background	II Good	III Moderate	IV Bad	V Very bad
	Background levels	No toxic effects	Toxic effects following chronic exposure	Toxic effects following short- term exposure	Severe acute toxic effects
As [mg/kg]	<20	20 - 52	52 - 76	76 - 580	>580
Cd [mg/kg]	<0.25	0.25 - 2.6	2.6 - 15	15 - 140	>140
Cr [mg/kg]	<70	70 - 560	560 - 5900	5900 - 59000	>59000
Cu [mg/kg]	<35	35 - 51	51 - 55	55-220	>220
Hg [mg/kg]	< 0.15	0.15 - 0.63	0.63 - 0.86	0.86 - 2	>2
Ni [mg/kg]	<30	30 - 46	46 - 120	120 - 840	>840
Pb [mg/kg]	<30	30-83	83 - 100	100 - 720	>720
Zn [mg/kg]	<150	150 - 360	360 - 590	590 - 4500	>4500

Figure 1. Location map with study area marked by black rectangles in A) regional and B) local context. Crossed hammers in B indicate the location of Ulveryggen mine. C) Map of Repparfjorden showing the locations of coring stations (for details see Table 1). Black circles on C indicate sediment cores for which results are presented within this manuscript; results for cores reflected with black circles/white fill are provided in Supplementary Figure 1 and 2. Green star indicates location of sediment core SED kjerne from Christensen et al. (2011). Dashed line marks a sill that separates inner from outer fjord. Red arrow indicates the probable position of pipeline outlet through which mine tilings were discharged in the 1970's.



Figure 2. Sedimentological and geochemical properties versus depth of eight sediment cores (for locations see Figure 1 and Table 1). Grain-size composition (clay – light, silt – medium and sand – dark grey). Total organic carbon (TOC) content.Heavy metal (As, Ba, Cd, Co, Cr, Cu, Hg, Ni, Pb, Ti, V and Zn) concentrations (upper x-axis); the color scale is in line with the Norwegian marine sediment quality classification of contaminants in marine sediments (see Table 7) (SFT 2007) (gray color is applied for metal concentrations not included in this classification); calculated values of geoaccumulation index (I_{geo}) (lower x-axis) after Müller (1969) where values of classes 0-1 are shown as dotted lines and classes >1 as solid lines. Pink horizontal bars mark the period of mine tailing disposal (1972-1978) based on the ²¹⁰Pb- derived and recalculated sediment accumulation rates (SARs) whereas the pink shading represents maximum and minimum ²¹⁰Pb- and ¹³⁷Cs-derived SARs (see Table 5).



Continuation of Figure 2.



Figure 3. Textural classification of all analyzed samples from all sediment cores (bottom panel) presented as sand-siltclay triangular diagram (after Folk 1954). Cores taken from different parts of Repparfjorden are marked by the following symbols color: white – inner fjord, light grey – northeastern part of outer fjord and dark grey – southwestern part of outer fjord.





Figure 4. Frequency curves of grain-size distributions of eight selected sediment cores (for location see Figure 1 and Table 1).

Figure 5. ²¹⁰Pb (upper x-axis) and ¹³⁷Cs (lower x-axis) activity profiles of eight selected sediment cores (for location see Figure 1 and Table 1). The horizontal error bars mark the 2-sigma measurement uncertainties.



Figure 6. Cluster analysis of 261 sediment samples of all investigated cores based on their sedimentological and geochemical signatures (for details see section 5.1). The smaller the linkage distance the bigger similarity between the samples. Distinguished clusters and sub-clusters (A, B1, B2 and B3) marked in gray shading (top and bottom panel) refer to sediment types discussed in the main text.



Supplementary Data Click here to download Supplementary Data: Sternal et al_SupplementaryTables.docx Supplementary Data Click here to download Supplementary Data: Sternal et al_SupplementaryFigures.docx