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An ecotoxicological assessment of mine tailings from three Norwegian mines

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HIGHLIGHTS

- Differences in particle and waterborne toxicity between the 3 mine tailings.
- Sibelco tailings were the most toxic based on waterborne exposures.
- Elevated metal concentrations in the Sibelco tailings were responsible for the toxicity.
- Fine particles (Hustadmarmor) showed highest toxicity when in contact with sediment.

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ABSTRACT

The study assessed the environmental toxicity of three Norwegian mine tailings from Omya Hustadmarmor, Sydvaranger, and Sibelco, which are all released into a seawater recipient. Ecotoxicity assessments were performed on the overlying water extracted from the mine tailings, the transformation/dissolution waters obtained from the mine tailings, and whole sediment assessment using a suite of marine organisms including algae, Crustacea, and Mollusca. Overall, based on the toxicity evaluation of the transformation/dissolution data, Sibelco tailings resulted in the highest toxicity albeit at relatively high concentrations, followed by Sydvaranger and Hustadmarmor. Sibelco was the only mine where process chemicals were not used. In contrast, the *Corophium* sediment contact assay revealed a significantly higher toxicity exerted by Hustadmarmor tailings, which may indicate a physical impact of the fine tailings. The effects observed were discussed with respect to both the measured chemical concentrations of the tailings and the potential physical impact of the tailing particles on organism health.

1. Introduction

The discharge of mine tailings into the sea is a relatively common practice in Norway, a situation that is not seen so frequently throughout the world. The potential benefits to this practice includes: the reduced cost of land transport and disposal; the redevelopment of harbour areas by reclaiming land from the sea (e.g. Bøkfjord, Kirkenes, Norway); and as a remediation tool for the capping of contaminated fjord sediments polluted with legacy contaminants such as PCBs and dioxins. Conversely, depositing large volumes of mine tailings into coastal ecosystems is a controversial subject and a concern for both scientists, regulators, and residents. The main concerns are over the physical and chemical impacts of the tailings on the local marine ecosystem, with potential harm to local fisheries and indirect impacts on public health.

The immediate impact of the tailings within the seawater recipient includes the physical smothering of benthic organisms near the discharge outlet, often referred to as the impacted zone. High turbidity is found within the impacted zone, which at high concentrations can have adverse effects on both feeding and respiratory organs of fish and invertebrates (Wilber and Clarke, 2001). Particle characteristics of the tailings strongly influence the intensity of the physical effects, which differ between mines. Furthermore, chemicals released within the tailings, either as naturally occurring compounds/elements from the mined rock or as added process chemicals introduced to the tailing slurry, to assist in the separation process, can have significant effects on organism health (Brooks et al., 2015; Brooks et al., 2018). The naturally occurring compounds/elements are mostly metals from the mined

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rock, whilst the process chemicals can include flocculating and flotation chemicals or other industrial chemicals, which may be released with the spent mine tailings. The chemical and physical properties of mine tailings can vary widely between individual mines and a comprehensive characterisation of the properties and biological effects of any mine tailing needs to be assessed before they are permitted for discharge into the sea. Ecotoxicity bioassays provide valuable information to understand the hazards that the mine tailings may pose to the environment. Such hazard data can subsequently be used to perform environmental risk assessments on the mine tailings prior to discharge.

The present study investigated mine tailings from three different Norwegian mines, with consents to discharge their tailings into three different Norwegian fjords. Evaluation of the potential direct and indirect effects of these mine tailings on marine organisms was performed using a battery of test approaches. These included, standard ecotoxicity assessments following internationally accepted test standards (e.g. ISO or ASTM) and through using targeted bespoke test designs. The test species used were native to temperate marine waters and representative of species that may be found in Norwegian fjords.

2. Methods

2.1. Mine tailings

The three main tailings that were assessed for their toxicity to marine organisms were from Norwegian mines that discharge their tailings into a seawater recipient (see Fig. 1 for approximate locations of the mines). Omya Hustadmarmor AS, Elnesvågen, located on the North-west coast of Norway in the county of Møre and Romsdal, receives marble from an open pit mine in Brønnøy in the county of Nordland. The marble is processed to obtain liquid marble for paper production, while the discharge tailings, containing mostly calcium carbonate with some quartz, feldspar, mica, and iron sulphide, are released into the Fraenfjord. These tailings can be categorised as fine grained; 0% > 63 μm, 40% > 20 μm and 70% > 4 μm (Arnstein Amundsen, Omya Hustadmarmor, pers.comm.). In addition to natural metal concentrations, the tailings contain both flotation (FLOT 2015, fictive name) and flocculation (FLOCC2014) chemicals used in the mining process.

Fig. 1. Location of the three Norwegian mines.

Sydvaranger mine, located in Kirkenes, Finnmark county, is an iron ore mine that between 2009 and 2015, discharged its tailings into the Bøkfjord. The tailings contained minerals such as quartz, amphibols, feldspar and metals in addition to the flocculation chemical Magnafloc (PolyDADMAC and polyacrylamide). Approximately 55% of the tailing particles were <63 μm (Trannum et al., 2018).

The Sibelco mine is located near Alta, also in the county of Finnmark, Northern Norway. This mine produces nepheline syenite and the discharge tailings contain natural metals as well as minerals such as amphibole, feldspar, nepheline, pyroxene, and biotite. No flotation or flocculation chemicals are added in the processing of this mine. According to Norwegian Mineral Industry (2014) about 45% of the tailing particles are <63 μm and 15% < 20 μm.

The mining companies supplied the tailings. The tailings were collected from each mine at the point in the process immediately prior to discharge into the fjord. The tailings were transported in large plastic containers (1000 L) to the NIVA Marine research station in Solbergstrand near Oslo. Homogenated subsamples of these mine tailings, obtained by thorough mixing with a glass rod, were used for testing.

2.1.1. Characterisation of tailing shape and size

Tailings obtained from the three mines were freeze dried for 24 h to remove any moisture from the samples prior to surface analysis. Morphological and local chemical information of the sediments were obtained by Scanning Electron Microscopy (SEM) coupled to Energy Dispersive X-Ray analysis (EDX). For these analyses a Hitachi S-3000 N Electron Microscope coupled to an EDX Bruker Esprit 1.8 unit was employed.

2.2. Assessment of the overlying water

Overlying water was only present in the mine tailings from Hustadmarmor and Sydvaranger as Sibelco was supplied as a dry tailing. Therefore, the toxicity assessment of the overlying water was only performed for these two mine tailings.

A toxicity identification evaluation (TIE) approach was used on the two overlying water samples to determine the group(s) of chemicals responsible for any observed toxicity (if present). To remove large particles, the overlying water was filtered through a 0.22 μm cellulose filter and the salinity was adjusted to 35‰ with a sea salt mixture (Instant Ocean). One part of the filtered salinity adjusted sample was placed through an SPE column, which removes organic chemicals from the water, whilst a second part was placed through a cation exchange (CE) column for the removal of metals from the samples. Subsequently, the toxicity of the prepared solutions, including the original overlying water, the SPE filtered and the CE filtered seawater from Hustadmarmor and Sydvaranger were assessed using the oyster embryo bioassay (ASTM E724-98, 2012). A 100% solution was used to represent the original overlying water concentration and 56, 32, 18, 10, 5.6, 3.2 and 1.8% dilutions of the original concentration were prepared with natural filtered seawater obtained from the outer Oslofjord. Filtered seawater was used as the control group. In addition, chemical analysis of the different elements present in the overlying waters were quantified using either Inductively coupled plasma mass spectrometry (ICP-MS) or Inductively coupled plasma atomic emission spectroscopy (ICP-AES).

2.2.1. Oyster embryo bioassay

The oyster embryo bioassay was performed in accordance with the ASTM guideline E724-98 (2012). Conditioned oysters were obtained from the Guernsey Sea farms, UK, and transported by overnight courier to the NIVA laboratories in Oslo, Norway. Oysters were processed within 1 h of receipt. Female oysters were opened with an oyster knife to reveal the internal tissues, rinsed briefly in filtered seawater before the gonad was pierced carefully with a glass pipette and stripped of eggs. The eggs were immediately placed in filtered seawater to create an egg suspension. The egg suspension was filtered through a 95 μm mesh, to remove large particles and clumps, and retained on a 25 μm mesh. The filtered eggs were then rinsed into a beaker and made up to a density of 3000 ± 300 eggs per ml with filtered seawater. The density of the egg suspension was determined through visual observations on a microscope (×40 magnification) using a 1 ml Sedgewick rafter cell. Male gametes were obtained in a similar fashion, although the sperm suspension was rinsed through a 60 μm mesh. The addition of sperm to the egg suspension in a ratio 1:200 ensured sufficient fertilisation of the eggs. The fertilised eggs developed into trochophore larvae after 2 h and these were used to inoculate the test vessels at a final density of 50 larvae per ml. The successful development of the oyster from trochophore to veliger larvae at 24 ± 1 °C for 24 ± 2 h was assessed microscopically (×40 magnification).

2.3. Transformation/dissolution experiments

Transformation/dissolution (T/D) experiments were performed on the three mine tailings according to methods described in Lillicrap et al. (2014) and OECD guidance document (OECD, 2001). For each tailing, 100, 10, 1 and 0.1 mg (dry weight) were mixed with 1 L of filtered (0.2 μm) seawater within a nitric acid rinsed plastic bottle. In order for the tailings to be added to the experiments on a dry weight basis, the wet to dry weight ratios of all three tailings were calculated. A 5 g sub-sample of each tailing was dried in an oven overnight at 80 °C until the weight remained stable. The wet to dry weight ratios of each tailing were used to adjust the amount of tailing added in the exposure studies.

The seawater that was added to the tailings was pH 8 and pH 6.2, creating two series of mine tailing T/D solutions, seawater without tailings was used as control. The natural pH of the seawater was 8, whilst a second volume of seawater was pH adjusted to 6.2 with 10 M HCl.

The filled bottles with secured lids, were shaken rigorously first by hand for 1 min, and then placed on an automated shaker (100 rpm) at 20 ± 1 °C. After 7 days, half of the bottles, one from each treatment concentration, at both pH, were removed and allowed to stand for approximately 6 h. After this time the overlying water was filtered (0.2 μm). Part of the filtered solution (ca. 15 ml) was analysed for different elements using ICP-MS and ICP-AES. The remaining solutions were used to determine the acute toxicity with the oyster embryo, the copepod *T. battagliai*, and a marine algal growth inhibition test. The remaining bottles were maintained under the same condition on the automated shaker for a total of 28 days after which time, the same procedure for the 7-day T/D was carried out.

2.3.1. Marine algal growth inhibition test (Skeletonema pseudocostatum)

The marine algal growth inhibition test was performed in accordance with ISO 10253. In brief, the test concentrations were inoculated with approximately 5×10^6 cells/L (algal density confirmed by coulter counter) of an exponentially growing culture of *Skeletonema pseudocostatum* (NIVA strain). Three replicates of each concentration were incubated in 30 ml glass vials with approximately 15 ml test volume on an orbital shaker at 22 ± 2 °C, under continuous illumination. Six replicate cultures in seawater only were used as controls. Growth of the controls was monitored daily using a Beckman Coulter Multisizer 3 to count cell number. Due to particle interference in cell counts at higher concentrations using the Coulter counter, a fluorimeter (Cytofluor 2300) was also used to monitor growth in all test vessels after 48 and 72 h.

2.3.2. Tisbe battagliai acute toxicity bioassay

Acute ecotoxicity assessments with the marine copepod *Tisbe battagliai* were performed in accordance with ISO 14669. The test animals were copepodid stage *T. battagliai* 6 ± 2 days old from in house cultures at NIVA. The test was performed in 12 well plastic plates and each concentration from each mine tailing had four replicates, each containing 5 *T. battagliai*. Approximately 4.5 ml volume per replicate well was used. The exposure vessels were placed in a climate controlled room (20 ± 2 °C) with a 16 h light: 8-h dark photoperiod. At 24 h intervals, observations were made for mortality/survival. Mortality is defined as the absence of any movement by the organism when examined with the aid of a microscope for 15 s. Dead animals were removed using a pipette. The test animals were not provided with food during the study.

2.4. Sediment contact tests

2.4.1. Oyster spat sediment contact assay (OSSCA)

Oyster spat (8 ± 2 mm) were provided by Guernsey Sea Farms (UK), and were exposed to the three individual mine tailings at concentrations of 0.1, 1, 10 and 100 g/L (d.w.). The tailings were housed in glass jars with lids and 250 ml of overlying filtered seawater. The tailings were allowed to settle for 5 days prior to the introduction of the oysters. The overlying water was aerated throughout the exposure. Ten oyster spat were placed in each test chamber with four replicates per treatment concentration. The length (mm) and weight (mg) of each oyster spat was recorded at the start of the test and again after 21 days when the test was terminated. Oyster spat were fed daily with live cultures of *Skeletonema pseudocostatum* at a density of 3000 cells per ml. The assay was performed in a constant temperature room at 20 ± 1 °C, salinity 35‰ with 16: 8 h light: dark photoperiod. Partial (50%) water exchange was made on day 5, 8, 12 and 15 of the 21-day test.

2.4.2. Corophium sediment contact assay

The sediment contact assay using *Corophium* sp. was performed based on the ICES Times 28 protocol but with some differences (Thain and Roddie, 2001). Live *Corophium* sp. and sediment were collected from a known clean reference location in the Trondheimsfjord and transported by courier to the NIVA laboratory in Oslo. The animals were acclimated for 5 days in filtered seawater at 15 ± 2 °C prior to testing. The tailings from the three mines were individually combined with reference sediment on a dry weight basis to achieve a tailing concentration gradient of 80, 50, 32, 18 and 1% of the original concentration. In 500 ml glass beakers, the tailings were mixed into a homogenous slurry using a glass rod and a small volume of the filtered seawater. Each treatment vessel contained 100 g of sediment/tailing homogenate with 300 ml of overlying filtered seawater. The treatments were settled for 3 h before the addition of 7 animals into each test vessel. Duplicates were used for each test treatment and control. The overlying water was aerated throughout the test and airlines checked daily. The bioassay was performed at 15 ± 1 °C with a seawater salinity of 36‰, pH 7.83–8.05 and DO 6.54–7.58 mg/L. The animals were not actively fed during the experiment. The test was terminated after 10 days and the number of surviving animals recorded.

2.5. Statistical analysis

Where homogeneity was measured with the Levene's test, the significant differences between the groups were compared using a one-way ANOVA and Tukey test ($p < 0.05$). When homogeneity was not achieved a non-parametric Kruskal Wallis test was used. ToxCalc 5.0. Scientific software was used to calculate toxicity NOEC, LOEC and EC_{50} values for the oyster embryo and the *Corophium* bioassays.

3. Results

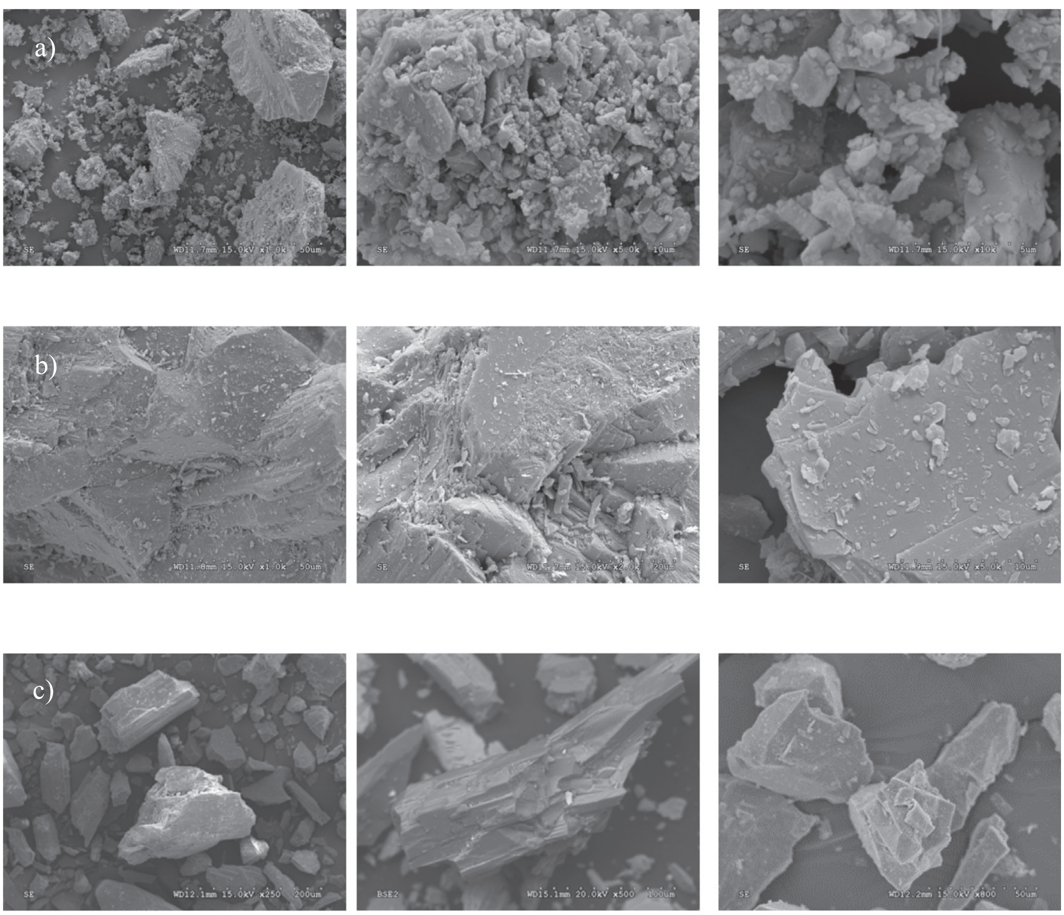
3.1. Characterisation of tailing shape and size

The sediments were morphologically characterized by SEM, and their surface composition analysed using EDX (Fig. 2). Tailings from Hustadmarmor consisted mainly of small particles with a large content of calcium. The shape of these particles in this tailing was rounded with relatively soft edges. Approximately 97% of Hustadmarmor particles were within the fine fraction (<63 μm) with a total organic carbon (TOC) of 6.4 μg/mg (Trannum et al., 2018).

The tailings from Sibelco and Sydvaranger contained marked amounts of both, Si, Fe, and Al. One important difference between the tailings was that those from Sibelco and Sydvaranger had particles with clear exfoliating joints compared to Hustadmarmor. The presence of these joints in the particles lead to the formation of fragmented and sharp-edged microparticles when placed under compressional forces and/or shear stress, which is expected during the mining process of mechanically crushed rock. Sibelco and Sydvaranger had 41% and 40% of particles within the fine fraction (<63 μm) with TOC <1.0 (Trannum et al., 2018).

3.2. Assessment of the overlying water using TIE approach

Since the Sibelco tailings were supplied by the mine in dry form, the overlying water was only available for testing from Sydvaranger



	% abundance							
Mine	**C**	**O**	**Mg**	**Al**	**Si**	**K**	**Ca**	**Fe**
HM	34.99	53.59	0.20		0.21		11.02	
SIB	14.45	43.10	1.29	1.06	11.92		0.66	27.53
SYD	47.20	42.19	0.85	0.74	6.12	0.20	0.84	1.86

Fig. 2. Scanning electron microscope (SEM) pictures at three magnifications and EDX analyses of element abundance (%) of the tailings from: row a) Hustadmarmor, row b) Sibelco and row c) Sydvaranger. Inserted table duplicated from Trannum et al. (2018).

and Hustadmarmor tailings. The overlying water from Hustadmarmor showed no toxicity to the developing larvae (data not shown). Consequently, the investigative treatment approach of the SPE and CE columns was not necessary.

For Sydvaranger, the toxicity of the overlying water to the development of the oyster embryo is shown in Fig. 3. Toxicity, which equated to a 20% reduction in normal embryo development, was only experienced at the highest concentration with a lowest observable effect concentration (LOEC) of 100% original overlying water. Treatment of the overlying water by filtering through an SPE column, and retesting, showed no difference from the untreated exposure. This would suggest that the toxicity observed at 100% concentration was not removed by the SPE columns. In contrast, enhanced toxicity was observed in oyster embryos exposed to the CE filtered overlying water with a LOEC of 32% of the original overlying water concentration. This response was contrary to that expected and was likely to be due to the removal of the essential ions from the media required by the embryos for normal development, rather than a toxicity response.

3.2.1. *Chemical concentrations of the overlying water*

The overlying water from Hustadmarmor and Sydvaranger tailings were measured for 33 different metal ions by ICP-MS. Since the Hustadmarmor tailings were freshwater and the Sydvaranger tailings were brackish (18‰), natural differences in the ion composition were evident. However, after normalising for seawater ions, eight compounds were found to dominate the two overlying waters (Table 1). For Sydvaranger, the dominant metals included Fe and Al with smaller contributions from Si, Mn and Cu. In contrast, Ba was the dominant metal in the Hustadmarmor overlying water, with smaller contributions from Fe, Al and Mn and less contributions from Si and Cu.

3.3. *Transformation/dissolution experiments*

The transformation dissolution (T/D) solutions made at pH 6 were at the lower limit of oyster embryo tolerance. As a result, 100% embryo abnormality was observed in the pH 6.2 control group. The oyster toxicity data for the T/D pH 6.2 was therefore not used.

Hustadmarmor had no sigificant effect on the development of the oyster embryos after exposure to either the 7 or 28 day pH 8 T/D solutions (NOEC>100 mg/L, Table 2). No effect on oyster development was observed when exposed to Sydvaranger 7 day T/D solutions, although toxicity was observed when exposed to the 28 day Sydvaranger T/D solutions (LOEC 100 mg/L). The highest toxicity to the oyster embryo was observed when exposed to the Sibelco T/D solutions, with LOEC values of 10 and 1 mg/L for the 7 and 28 day T/D solutions, respectively.

The effects of the T/D solutions on growth and yield in the marine algae *S. pseudocostatum* are shown (Fig. 4). Significant reductions in growth and yield were exhibited by all mines at even the lowest concentration of 0.1 mg/L d. w. (ANOVA, Tukey $p < 0.05$). The Sibelco T/D solutions showed the largest reduction in both algal growth rate and yield of approximately 80% of 35% respectively of the control value at 10 mg/L d. w. The growth rate and yield at the highest Sibelco concentration (100 mg/L d. w.) was approximately 90% and 60% of the control value respectively, and above the values at 10 mg/L d. w. Smaller changes in growth rate following exposure to Hustadmarmor and Sydvaranger where observed (approximately 90% of control), whilst yield was reduced to approximately 80% and 70% of control values for Hustadmarmor and Sydvaranger

Table 1
The dominant metal ions measured in the overlying waters of the two mine tailings.

Metal ion	Unit	Sydvaranger		Hustadmarmor		Seawater reference
Al	μg/L	69.11	65.33	10.79	11.38	1.54
Si	mg/L	5.76	5.76	2.02	2.11	0.23
Mn	μg/L	3.19	3.13	10.36	10.64	0.23
Fe	μg/L	151.00	141.33	20.19	22.06	5.48
Cu	μg/L	1.45	1.32	2.73	2.49	0.51
Ba	μg/L	5.86	6.24	102.43	107.30	7.21

Table 2
The effects of the 7 and 28 day transformation dissolution (T/D) exposure on the development of the oyster embryo after 24 h exposure (values in mg tailing per litre of seawater, pH 8). No Observable Effect Concentration (NOEC), Lowest Observable Effect Concentration (LOEC), Effect Concentration causing effects in 10% (EC10) and 50% (EC50) of the population.

Mine	T/D mixing duration (days)	NOEC	LOEC	EC10	EC50
Hustadmarmor	7	>100	>100	–	–
	28	>100	>100	–	–
Sydvaranger	7	>100	>100	–	–
	28	10	100	16.7	>100
Sibelco	7	1	10	1.39	7.86
	28	0.1	1	0.42	7.32

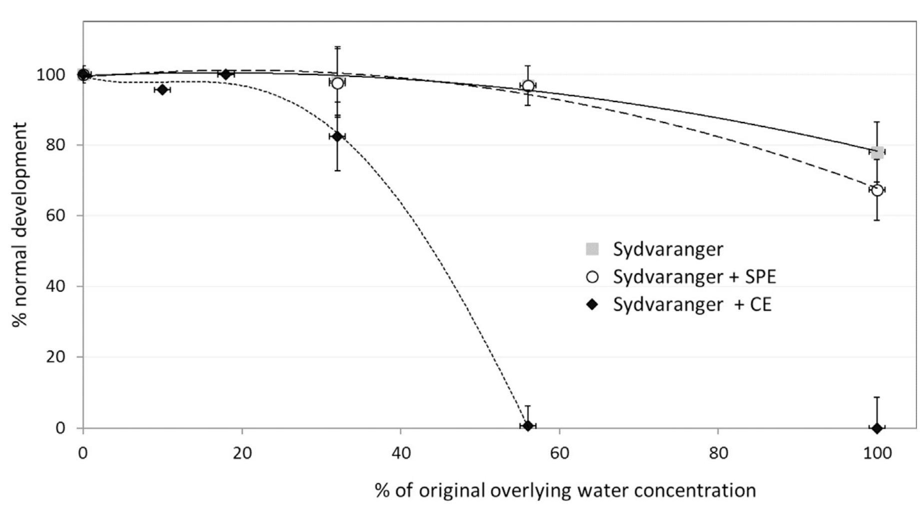


Fig. 3. Toxicity of the overlying water from Sydvaranger mine tailing to the development of the oyster embryo. Overlying water treated by solid phase extract (SPE) and cation exchange (CE). Mean ± standard deviation, n = 4.

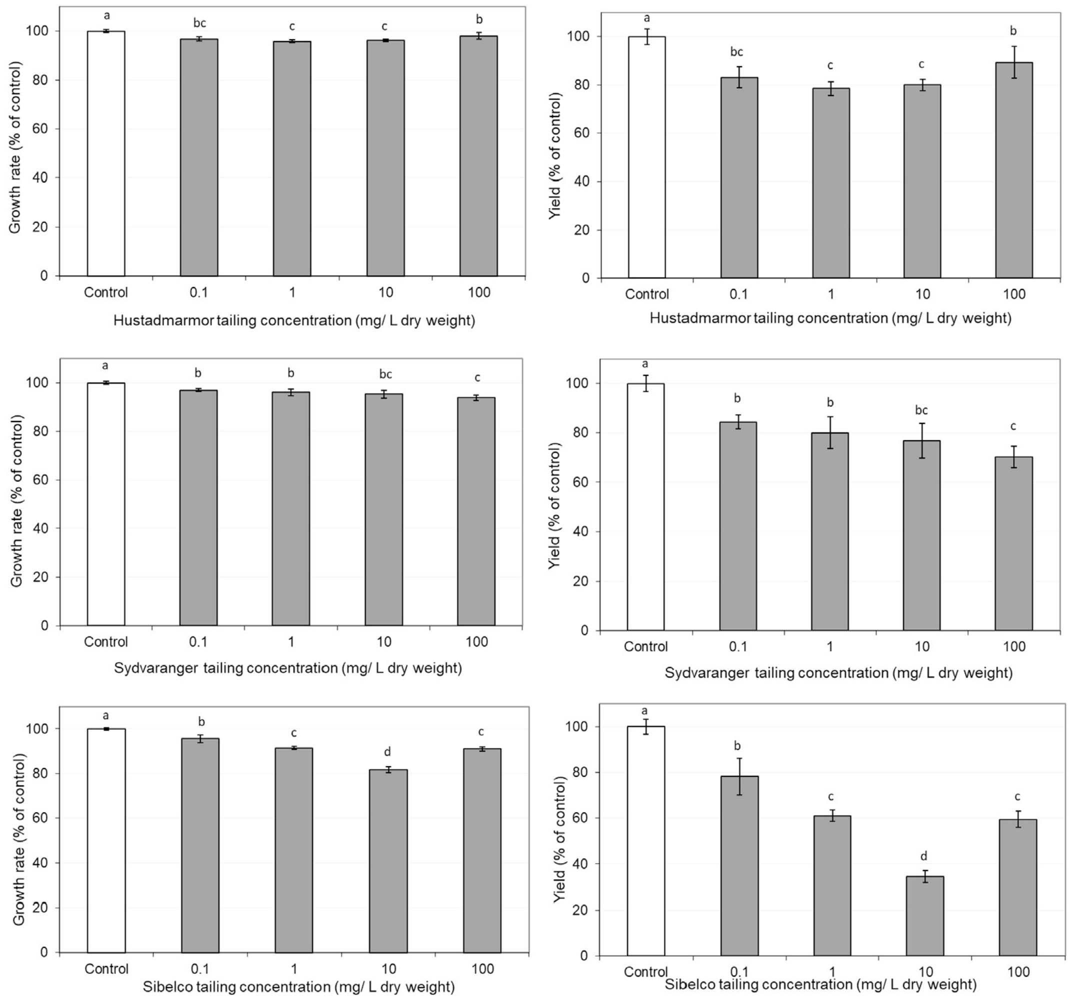


Fig. 4. The effects of the 7 day transformation/dissolution (pH 8) of the three tailings on the growth of the marine microalgae *Skeletonema pseudocostatum*. The tailing concentration relates to the amount of tailing (mg) added to 1 L of seawater before mixed for 7 days in the dark. Mean ± standard deviation, n = 5. Significant difference between treatment concentrations for each tailing denoted by different letter (ANOVA, Tukey $p < 0.05$).

respectively.

No acute toxicity was observed in *T. battagliai* after 48 h exposure to any of the three mine tailing T/D solutions (Data not shown).

3.3.1. Chemical concentrations of T/D solutions

The measurement of 33 metal ions in the T/D solutions of the three mine tailings was performed by ICP-MS and ICP-AES. Of these, eight metal ions were found to be markedly elevated above control seawater levels for at least one of the mines and was used to characterise the three tailings (Fig. 5). Sibelco tailings contained elevated concentrations of Al (132 μg/L), Mn (327 μg/L), Co (1.3 μg/L) and Ba (134 μg/L) in the dissolved fraction of the T/D exposure solutions, whilst Sydvaranger contained higher levels of Cu (4 μg/L), Mn (321 μg/L), Co (1.1 μg/L), Ni (9 μg/L), Cs (25 μg/L), Ba (248 μg/L), and Zn (7 μg/L) compared to control seawater. In contrast, Hustadmarmor contained concentrations of Cu (7 μg/L), Co (0.9 μg/L), Ni (9 μg/L) and Ba (103 μg/L) above the control seawater concentrations.

3.4. Sediment contact bioassays

3.4.1. Oyster spat sediment contact assay (OSSCA)

The effects of the three mine tailings on the growth of the juvenile spat of the Pacific oyster (*C. gigas*) after 21 days is shown in Fig. 6. For all three tailings, there were no significant differences between the control and the treatment groups after 21 days exposure.

3.4.2. Corophium bioassay

The effects of the mine tailings on the survival of the amphipod, *Corophium* sp. after 10 days is shown in Fig. 7. All tailings showed significant effects on *Corophium* survival at different concentrations. The Sydvaranger tailing showed least toxicity with a NOEC, LOEC and EC_{50} concentration of 50%, 80% and 130% of the original tailing concentration respectively. Sibelco was slightly more toxic to *Corophium* with a NOEC, LOEC and EC_{50} of 50%, 80% and 80.3% of the original tailing concentration respectively. However, Hustadmarmor was the most toxic tailing to the *Corophium* with NOEC; LOEC and EC_{50} values of 18%, 32% and 45.6% of the original tailing concentration respectively.

4. Discussion

The investigative TIE approach of the overlying water was to determine which chemical(s) were responsible for any observed toxicity to the developing oyster embryos. However, the lack of toxicity of the Hustadmarmor overlying water to the oyster embryos, which is one of the most sensitive marine bioassay species to

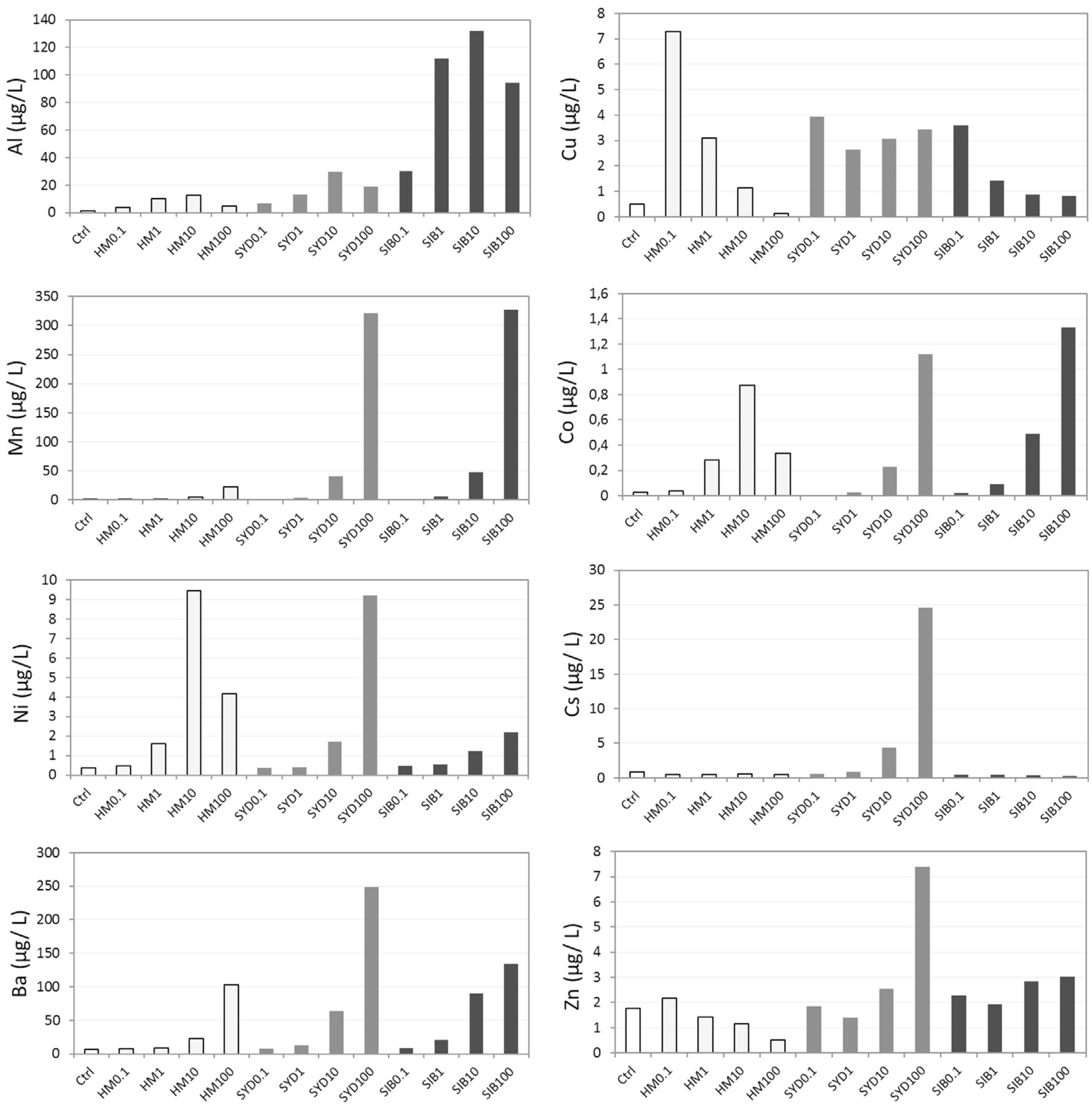


Fig. 5. Selected metal analyis from the prepared 7 day transformation/dissolution mixtures (pH 8) from the three main tailings, Hustadmarmor (HM), Sydvaranger (SYD) Sibelco (SIB).

metal toxicity (Brooks and Waldock, 2008), made the TIE approach redundant for this mine tailing. The overlying water from the Sydvaranger tailings did show some toxicity at the highest concentration when undiluted. The overlying water when filtered through an SPE cartridge showed an identical response and indicates that the organic compounds were unlikely to be responsible for the observed toxicity. It was more probable that metals within the overlying water and particularly Fe and Al, with contributions from Si, Mn and Cu, as measured by ICP-MS, contributed to the toxicity observed. The CE approach has been used to remove metal toxicity from environmental samples (Gerssen et al., 2009). However, in the present study the CE column did not remove the toxicity but rather enhanced it, and this was considered to be due to the removal of the essential ions in addition to the potentially toxic metals from the overlying water. It was therefore likely that metals present in the overlying water were responsible for the observed effects, although the investigative approach used was unable to scientifically confirm this.

Analysis of the T/D solutions revealed differences in the metal concentrations between the three mines. Sibelco tailings were dominated by Mn, Al, and Ba; whilst Hustadmarmor was dominated by Ba, Cu, and Ni; and Sydvaranger by Mn, Ba, and Zn. Although some toxicity was observed, the overall acute and chronic toxicity was low with only the undiluted tailing (100% concentration) causing a toxic response in developing oyster embryos. The assessment enabled toxic responses to be differentiated between chemical dissolved and particle effect toxicity.

The effects of the dissolved concentrations of metals from the different mine tailings showed that Sibelco was the most toxic of the three mines. Toxic responses were observed in both oyster embryo development and growth inhibition of the marine algae. Process chemicals were not used at this mine and the toxicity was most likely due to the combined effects of elevated concentrations of metals such as Al (100 µg/L), Mn (325 µg/L) and Ba (140 µg/L). From the United States Environmental Protection Agency (US EPA) toxicity database, the toxicity of Al, Mn, and Ba to marine organisms has been reported. A 72 h EC_{50} between 0.81 and 14.84 mg Al/L was found for the developing embryos of the grass shrimp (Rayburn and

Aladdin, 2003), whilst oyster embryos resulted in an EC_{50} of 16 mg/L for Mn (Nelson and MacInnes, 1973). However, Ba was the most toxic with an EC_{50} of 0.189 mg/L for developing mussel embryos (Spangenberg and Cheer, 1996) and may be a contributing factor to the toxicity from the Sibelco mine tailings.

Sydvaranger showed slight inhibition in algal growth and abnormal embryo development only when exposed to 28-day T/D solutions at the highest concentration. Flocculants are used at Sydvaranger to collect the silicate minerals and enable the process water to be recovered and recycled (Floer, 2015). The composition of the T/D solutions following the partition of chemicals from the tailings to the dissolved phase can be influenced by several factors including temperature and salinity. The presence of chemical flocculants in the sample, which are designed to aggregate the particles, may reduce the partitioning of chemicals such as metals to the dissolved phase (Karbassi and Nadjafpour, 1996). Since the T/D solutions are filtered prior to exposure in the bioassays, it is only chemicals in the dissolved phase that are exposed to the organisms. Therefore, the presence of flocculants in the tailings may be partly responsible for the relatively lower toxicity of the Sydvaranger tailings. However, the dominant metals in the Sydvaranger T/D solutions were Mn (321 μg/L), Ba (248 μg/L) and Cs (25 μg/L), with smaller contributions from Ni, Zn, and Cu. Interestingly the Ba concentration for Sydvaranger T/D solutions were higher than that observed for Sibelco and above the Ba EC_{50} found for the development of mussel embryos (Spangenberg and Cheer, 1996). However, although Sydvaranger T/D solutions exhibited a toxic response, the effects observed were relatively less than the Sibelco tailing T/D solution. The reason for the higher toxicity of the dissolved fraction of the Sibelco tailings is likely to be due to toxicity contributions from other chemicals that were present but not measured.

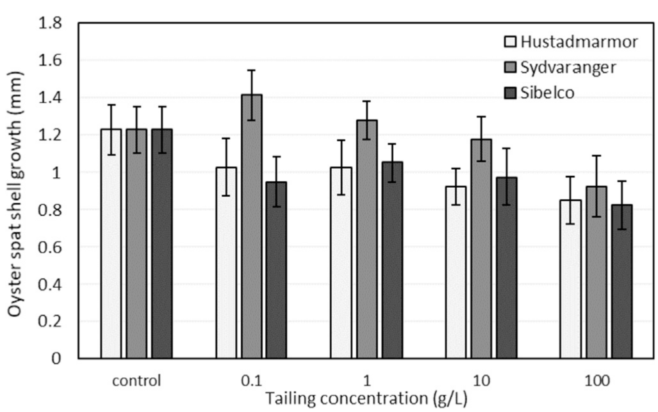


Fig. 6. The Oyster spat sediment contact assay: the effects of 21-day exposure to the three mine tailings on the growth of the oyster spat (*C. gigas*). (Mean ± SE, n = 40, no statistical difference between groups ANOVA, Tukey $p > 0.05$).

The overlying water or the T/D solutions from the Hustadmarmor tailings showed a significant but small inhibition to algal growth rate and yield. The metal analysis revealed the lowest concentration of metals of the three mines. Only Ba (100 μg/L) and Cu (7 μg/L) were slightly elevated but did not cause a measurable toxic response. Flotation chemicals (FLOT 2015) are known to be used within the processing of the tailings to optimise mineral recovery at this mine and are released with the tailings into the recipient. The detection of this flotation chemical has previously been reported in mussels positioned up to 2 km from the discharge outlet and indicates a widespread distribution upon release (Brooks et al., 2018). Hustadmarmor is a well-studied mine discharge and data on the impacts on the benthic ecosystem in the Fraenfjord recipient has been previously reviewed (Brooks et al., 2015).

Of the three tailings studied, it was the mine with no process chemicals added (Sibelco) that showed the greatest effects. Although surprising, it does support earlier studies where these mine tailings have been previously assessed. For example, low toxicity of Sydvaranger tailings containing the flocculant (Magnfloc) were reported in several species including the marine algae *Skeletonema costatum*, the lugworm *Arenicola marina*, the copepod *Tisbe battagliai* and the juvenile turbot *Scopthalmus maximus* (Berge et al., 2012, 2014).

The impact of particles from the sediment contact assays revealed a different response between the three mine tailings. Although Hustadmarmor showed no toxicity following exposure to the dissolved fraction of tailings, the sediment contact assay using *Corophium* sp. showed the largest toxic response of the three tailings. This may suggest that *Corophium* sp. were particularly

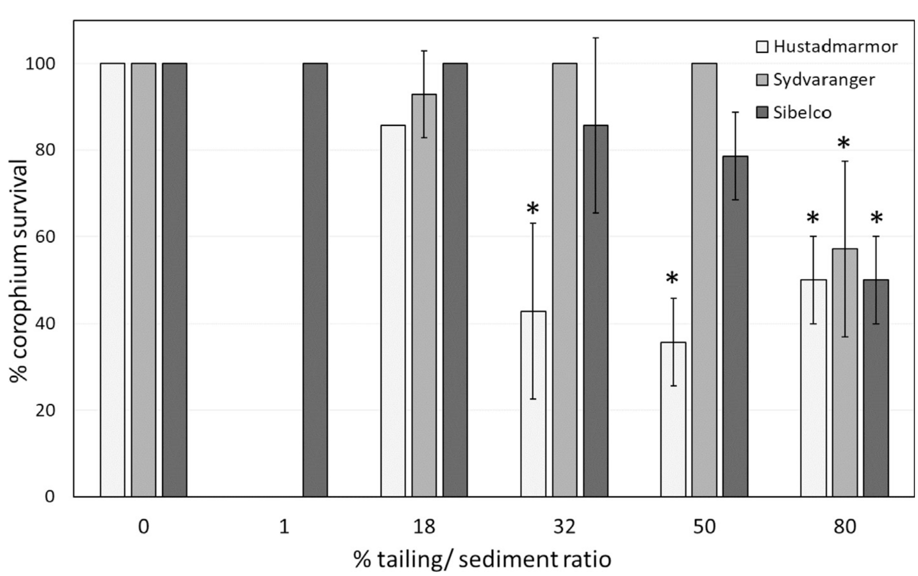


Fig. 7. The effects of a 10-day exposure to three mine tailings on the survival of the sediment dwelling *Corophium* sp. (mean ± SD) * significantly different from control ANOVA. Tukey $p < 0.05$).

sensitive to components of the Hustadmarmor tailing relative to the other two mines. However, this was unlikely to be a response to chemical exposure, since *Corophium* sp. would not be expected to be more sensitive then *T. battagliai* or oyster larvae as previously tested. The response observed is most likely due to the physical interference of the particles to the *Corophium* sp. Scanning electron microscopy (SEM) of the physical interference of the particles revealed that the Hustadmarmor tailings were relatively smaller with round soft edges. The Hustadmarmor tailings have been previously reported to consist of a fine material (<63 μm) as high as 97%, with Sibelco and Sydvaranger tailings comprising of 41% and 40% fine material (Trannum et al., 2018).

In contrast, both Sibelco and Sydvaranger had exfoliation joints, which resulted in platelets and needle-shaped microparticles (see Fig. 2). It was originally assumed that the sharp-edged needle like particles may cause increased damage to the exposed organisms through contact with sensitive external structures. However, it appears that the smaller and finer particles from Hustadmarmor had a greater impact, with possible particle interactions with gill filaments. The effects of natural mineral particles to rainbow trout gill epithelial cells *in vitro* revealed that particles < 2 μm in diameter were taken up by the cells enabling mineral species to interact with gill cells resulting in observable levels cytotoxicity (Michel et al., 2014). It was found that the fine silt and clay sized mineral particles where responsible for cytotoxicity in gill epithelial cells. It may be feasible therefore for the very fine particles within the Hustadmarmor tailings to be taken up by *Corophium* gill cells resulting in negative interactions.

The Sibelco and Sydvaranger tailings did show significant mortalities to *Corophium* sp. in the sediment contact assay at the highest concentration and this may suggest a particle interaction in addition to the chemical effects previously demonstrated from the T/D exposures. The SEM revealed that both Sibelco and Sydvaranger tailings had similar needle-like particle structures, and the observed similarity in biological response when exposed to *Corophium* may be a reflection of the similarity in particle form exhibited by these two mine tailings.

The relatively stronger effects of the Hustadmarmor tailings on *Corophium* sp. in the sediment contact test was also observed in a parallel study that investigated the impact of the same three mine tailings on benthic community structures (Trannum et al., 2018). Effects were observed in the different faunal feeding groups including surface deposit feeders and suspension feeders. The authors were surprised by the lack of effects observed by the Sibelco and Sydvaranger tailings compared to those from Hustadmarmor, particularly since Sibelco and Sydvaranger had sharp needle like particles, as previously described. Particle shape, such as those with sharp edges and needle-like projections have been shown to impact fish gills and induce fish stress responses (Lake and Hinch, 1999). Ingestion of sharp-edged mine tailings by the marine copepod *Calanus finmarchicus* was thought to have contributed to some of the adverse effects (Farkas et al., 2017). However, in all these examples, particle shape was not considered to be responsible for causing mortality in the exposed organisms but rather contributing to sub-lethal effects.

The physical and chemical impact of the tailings on the local marine ecosystem is an important consideration with respect to the direct discharge of tailings into a seawater recipient. Although it is difficult to directly relate toxicity observed in controlled laboratory studies to field scenarios, they can provide some indications of the potential environmental impact. Based on the observed toxicities from the overlying water and T/D solutions of the three mines, the impact on the marine environment would not be expected to be significant, particularly when considering high dilution factors within the receiving waters. Care should be taken however, since continuous mine tailing discharge into receiving waters that are less well mixed may create accumulation of chemicals creating contaminant hotspots. It is therefore important that suitable field monitoring in mine recipients is performed on a regular basis in order to measure potential impacts.

The particle effects observed from the Hustadmarmor tailings was thought to indicate a potential impact of fine particles on gill physiology. Information on the impact of fine natural mineral particles to marine life is limited, although research on nanoparticles would suggest that exposure to very small particles have their own additive effects in addition to chemical exposure (Handy et al., 2008). The fine Hustadmarmor tailings are known to cause high turbidity in the receiving waters of the Fraenfjord in Norway (Brooks et al., 2015). The interaction of fine mineral particles with gills of marine organisms is somewhat unknown. However, previous investigations have shown significant biological effects in the filter feeding mussel (*Mytilus edulis*) positioned within 2 km of the discharge outlet (Brooks et al., 2018). Determining the interactions of Hustadmarmor fine tailings on gill epithelial cells of marine organisms is recommended to determine if they pose a real threat to marine life.

5. Conclusions

The effects of the dissolved concentrations of metals from the three mine tailings showed that the tailings from Sibelco resulted in the highest toxicity, albeit at relatively high concentrations, followed by Sydvaranger and Hustadmarmor. Toxic responses were observed in both oyster embryo development and growth inhibition of the marine algae. Process chemicals were not used at Sibelco and the toxicity was most likely due to the combined effects of elevated concentrations of metals such as Al (100 μg/L), Mn (325 μg/L) and Ba (140 μg/L). The impact of mine tailing particles from the sediment contact assays revealed a different response between the three mine tailings, with Hustadmarmor showing the largest effects on *Corophium* sp. survival. Interference of the fine particles of Hustadmarmor with the gill epithelia of marine organisms was thought to be a contributing factor. This may have implications to marine organisms that occupy the high turbidity seawater recipient of the Hustadmarmor mine and studies investigating the interactions between fine mineral particles and gill epithelial cells of marine organisms are recommended.

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