Contents lists available at ScienceDirect





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



In situ characterisation of complex suspended particulates surrounding an active submarine tailings placement site in a Norwegian fjord



Emlyn J. Davies *, Raymond Nepstad

SINTEF Ocean, Environmental Technology, 7465 Trondheim, Norway

ARTICLE INFO

Article history: Received 27 March 2017 Received in revised form 14 September 2017 Accepted 17 September 2017 Available online 25 September 2017

Keywords: Flocculation Submarine mine tailings placement Particle size distributions Sediment transport Settling Particle imaging

ABSTRACT

Suspended particle measurements over flood and ebb transects of a Norwegian fjord (Frænfjorden) with an active submarine mine tailings placement are presented. Measurements focused on characterisation of spatial variability in particle and floc size, and determination of suspended mine tailing concentrations. Such information is crucial for understanding transport and flocculation dynamics, which in turn can contribute to improvements of numerical models, and thus to more accurate predictions of settling fluxes and transport processes. Multiple instruments were used to ensure that all relevant particle sizes could be observed. These included a LISST-100x, a LISST-Holo and a bespoke particle imaging system, which were deployed together on a profiling frame at multiple stations transecting the fjord. Measurements were obtained close to the high-concentration plume from the tailings discharge and extended several kilometres horizontally. We observed a combination of few-micron fines, densely packaged flocs and large string-like flocs, several mm in length; a range which could not be captured by a single instrument. Such a range of floc sizes and complexities has significant implications for the transport and settling of the material suspended within the fjord.

© 2017 The Authors. Published by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).

1. Introduction

Mining in Norway is currently in a phase of increased growth and subject to evolving environmental regulations and requirements. For many mines, both in Norway and globally, sustainable waste management remains the main challenge, as they often produce large quantities of waste rock and tailings from landbased mineral processing during resource extraction (Skei, 2013). To overcome problems with deposition of this waste, some mines place their tailings at the sea floor of fjords as submarine tailings placements (STPs) (Dold and Bernhard, 2014; Ramirez-Llodra et al., 2015; Skei, 2013). In Norway, there are currently 33 sites that have been identified as possible locations for STPs, of which 6 are active today (Ramirez-Llodra et al., 2015).

The best available techniques should be used to ensure that the environmental impact from STPs are reduced to an absolute minimum, and that ecosystems are given the chance to recover as quickly as possible following the closure of mines. The discharged mine tailings have the potential to affect sea bed ecosystems, through smothering, change of grain size or dissolved metals and chemical toxicity, as well as organisms living in the water column,

* Corresponding author.

E-mail address: emlyn.davies@sintef.no (E.J. Davies).

either through dissolved substance toxicity or from suspended fine-grained tailings (Doe et al., 2017; Morello et al., 2016; Tye et al., 1996). It has been long known that pelagic filter feeders can ingest mineral particles upon exposure (Anderson and Mackas, 1986; Kach and Ward, 2008), and more recently, it was shown that fine-grained mine tailings can also attach to the surface of a calanoid copepod species (Calanus finmarchicus) (Farkas et al., 2017). No acute effects were observed in that study, but longer term effects from the mine tailings could not be ruled out. The potential for environmental exposure of such organisms to fine grained tailings is therefore important to assess. In addition, large flocs can act as carriers of hazardous substances (Lyons et al., 2005) and increase the exposure of contaminants on the seabed due to the loosely-packaged sediment layer that forms from floc settling (Milligan and Law, 2013). Knowledge on how best to dispose of mine tailings in the marine environment to minimise ecological impacts requires a detailed understanding of exposure pathways and mechanisms of biological impact, which in turn requires a thorough understanding of the transport mechanisms.

A key process determining the transport and fate of mine tailings in the sea is aggregation, or flocculation, whereby small mineral particles collide with each other and with biological and mineral particles already present in the water, to form flocs of various shapes, sizes and densities. This alters the fate and transport of the discharged material by changing the settling behaviour

http://dx.doi.org/10.1016/j.rsma.2017.09.008

^{2352-4855/© 2017} The Authors. Published by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).



Fig. 1. Map showing the location of Frænfjorden (red rectangle). The inset shows a closeup with Frænfjorden and the STP area indicated (red rectangle in inset). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

of the particles (Hill et al., 2000; Skei and Syvitski, 2013). Two things change in relation to particle settling fluxes that result from flocculation: (1) increase in particle size; (2) decrease in particle density. The result is the formation of flocs which sink faster than the individual component particles (flocculi) but slower than would be predicted from solely the increase in size and no consideration of reduction in density (Mikkelsen and Pejrup, 2000; Graham and Nimmo-Smith, 2010b; Bowers et al., 2014). This means that some large flocs have the potential for significant horizontal transport if their density is sufficiently low. In order to understand the degree to which flocculation alters these transport processes it is necessary to characterise the size distribution and concentration of particles using methods that span a size range of several orders of magnitude. A single instrument or technique cannot obtain measurements over such ranges (Boss et al., 2015). Thus, a combination of instruments must be adopted (Mikkelsen et al., 2005; Reynolds et al., 2010).

Here we present detailed *in situ* measurements of the distribution of suspended mine tailings within Frænfjorden, Norway, where approximately 35 mt/h of dry mass material is discharged into the fjord in a solution of saline water with approximate drywet ratio of 8%. These measurements were targeted at obtaining information at multiple locations throughout the fjord, covering flood and ebb flows and intersecting the STP area. Detailed information about the nature of the particle populations present was also of interest in order to assess the degree to which flocculation may affect the fate of discharged material. This insight is necessary in order to improve our understanding of environmental consequences within the pelagic zone, and ultimately within the benthic zones following sedimentation.

1.1. Study site

The STP site investigated is located in Frænfjorden, near the city of Molde, Norway (see map in Fig. 1). Owned by Omya Hustadmarmor AS, the STP has been in operation since the early 1980s. The main component of the discharged tailings is calcium carbonate from limestone production, with a significant fraction of very fine particles (31% less than 2 μ m, and 71% less than 20 μ m. Before discharge, the tailings, which contain some residual fresh water,



Fig. 2. Detailed map of Frænfjorden, showing the location of the release point during the measurement period (red arrow), the acoustic current profilers on bedframes (red markers), and the profiling stations (green markers). The size of the profiling station indicators are proportional to the maximum turbidity at that location. eastings and northings are given in UTM zone 32 V coordinates. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)



Fig. 3. Photograph of the profiling frame during recovery at the end of a profile. A white cloud of mine tailings can be seen surrounding the frame, as the accumulation of material collected on the frame 'feet' at the seabed, is released.

are mixed with sea water for an approximate final concentration of 8% solids. For more information, see Ramirez-Llodra et al. (2015).

Frænfjorden is a narrow and shallow fjord, connected to the larger Romsdalsfjord on the outside (Fig. 1 inset). The main part of the fjord extends east–west about 7 km and is about 1 km wide. Depths are mostly less than \sim 70 m in the inner part of the fjord. The mine tailings discharge was located at UTM coordinate 404 000 E, 6 969 600 N, and a depth of 20 m at the time of this study (June 2015).

2. Method

Two acoustic current profilers were deployed on bedframes, one close to the main inlet to the fjord, and one close to the release point (red markers in Fig. 2). These provided a three-day timeseries of water column currents at each location.

A suite of instrumentation for quantifying the size distribution and concentration of suspended particles was deployed from a profiling frame (Fig. 3) along flood and ebb transects, comprising a total of 33 profiling stations. Each station where profiles were obtained is plotted as a green marker in Fig. 2. Turbidity – a relative measure of the optical clarity of the water – was measured



Fig. 4. Photograph of the instrumentation mounted on the profiling frame. The LISST-HOLO, LISST-100 and SilCam were mounted such that the sample volumes were unaffected by wake from the frame during profiling.

using an OBS (optical backscatter sensor) attached to the CTD (Conductivity, Temperature, Depth). The size of the green markers in Fig. 2 is proportional to the maximum measured turbidity (plus a minimum size offset). Particle size distributions (PSDs) and concentrations over the range of $5-8000 \,\mu\text{m}$ were measured using a combination of the LISST-100x, LISST-Holo, and a bespoke particle imaging system (called SilCam for convenience) designed for measurements of large particles in high concentration (Davies et al., 2017). These instruments (Fig. 4) were selected for their ability to measure suspended material *in situ* with minimal disruption to the particles being measured. This is of critical importance when studying flocculated materials because flocs are sensitive to turbulent shear (Hill, 1998).

The LISST-100x is a Laser *In Situ* Scattering and Transmissometer, which uses the principles of forward-angle light scattering to estimate the size distribution and concentration of particles (Agrawal and Pottsmith, 2000). The instrument can make this quantification in the size range of $2.5-360 \mu$ m, using the nonspherical inversion matrix (Agrawal et al., 2008).

The LISST-Holo is an *in situ* holographic camera, which uses inline digital holographic imaging (Owen and Zozulya, 2000; Graham and Nimmo-Smith, 2010a) to obtain in-focus particle images at all depths along the 5 cm path length. The system can quantify particles within a range of $30-2000 \,\mu$ m in diameter.

The SilCam imaging system (Davies et al., 2017) uses the principles of transmittance and silhouette imaging, together with telecentric optics, to obtain in-focus particles within a 10 mm path length. The system can quantify particles over the range of 56-8000 µm in equivalent circular diameter. The equivalent circular diameter is the diameter of a circle of equal area to the particle in question, and is used as a metric for size measurements when imaging particles of a wide variety of shapes. It is worth noting that this metric for estimating particle size is based on a projected area, and can therefore be subject to some errors in cases where elongated particles are seen end-on such that the longest axis is not visible. However, this error becomes reduced for in-situ measurements where particle orientations are often random. The SilCam system has a large imaging area (35.2 mm \times 29.4 mm) and high sampling frequency (up to 15 Hz) in comparison to the LISST-Holo. This gives reliable statistics for the particles larger than 2 mm, where the LISST-HOLO image area and low acquisition

frequency starts to become limiting when used as a profiling instrument. The volume of water quantified is therefore a combination of the instrument sample volume and acquisition frequency. Typical volumes of water sampled per profile were: \sim 6 L/profile, \sim 0.02 L/profile, and \sim 15 L/profile for the LISST-100x, LISST-Holo and SilCam, respectively.

The choice of a 10 mm path length for the SilCam system was made to enable measurements of high concentrations of large particles close to the release point. The LISST-100x and LISST-Holo were both deployed with standard 50 mm path lengths to quantify small particles in lower concentration further away from the release site. As optical backscatter is also primarily sensitive to small particles (Davies et al., 2014), the gain setting of the OBS was selected to obtain greater detail of the lower turbidity regions, as opposed to the area close to the release.

3. Results & discussion

Data from the two current profilers show that the main circulation within the fjord is controlled by a diurnally-asymmetric, tidally-driven flow (see Fig. 5). The circulation close to the release point was harder to interpret, likely due to a more complex flow regime within this area of the fjord.

Fig. 6 shows the variation in turbidity with depth for all stations, separated by ebb and flood tide and projected onto an East– West grid. Density contours are shown together with the turbidity colourmaps, with depth- and location-averaged values shown in the outer plots. Three key features are evident in the spatial variation in turbidity: (1) The highest turbidity is found directly below the release point, where the OBS response saturated; (2) The upper extent of the plume of discharged material was always below the pycnocline (at approximately 8 m depth) and often several metres below the pycnocline; (3) There exists an area of high turbidity to the East of the release point below depths of approximately 25 m. These three observed features are persistent over both flood and ebb measurements, thus implying some degree of tidal pumping or residual circulation that acts to retain the material to the East of the release.

In the high-concentration area close to the release (possibly inside the plume), the median particle diameter (d_{50} of the volume distribution) was approximately 300 μ m. This area was too concentrated for reliable LISST-100 measurements without the path reduction module (as the transmission through the instrument path length was less than 40%), and was also sufficiently concentrated to saturate the OBS. However, due to a dominance of relatively large particles, this concentration was within a reliable limit for the SilCam system with the shorter, 10 mm path length.

Fig. 7 shows the total volume concentration of particles of 56– 360 μ m, as measured by the SilCam. The trends seen in the SilCam and turbidity are consistent, with the exception of areas where there is a dominance of small particle concentrations (supported by LISST-100x observations). In these areas turbidity shows a signal that is not resolved by the integration of the 56–360 μ m range of the volume distribution.

Some of the results from LISST-100x occasionally contained features that were difficult to explain, and also inconsistent with turbidity and the SilCam. However, using the *in situ* image data, it was possible to perform quality control measures on the LISST-100x data. For example, regions of contamination due to Schlieren (optical inhomogeneities, mostly around the pycnocline at 8 m) were identified in images from the LISST-Holo and SilCam, and were discarded from the LISST-100x data (see Mikkelsen et al., 2008 for further details), and two profiles were discarded due to fouling of the optical windows.

From the point of release, a high concentration of initially small particles are dispersed in the form of a dense plume, which take approximately 60 s to fall the 20 m distance from the release point to



Fig. 5. Velocity components measured by the current profiler placed at the fjord entrance.



Fig. 6. Coloured contour map of turbidity along ebb (A–C) and flood (D–F) transects, with depth and location averaged data shown in the outlying plots. The outlying plots show depth-integrated (A, D) and horizontally-integrated (C, F) values, where individual observations are shown by grey markers, with the total averaged values shown by the black lines. Black contours in B and E are isopycnals, and the thick black line shows the seabed depth along the transect. The position of the release point for the discharge is shown by the yellow squares (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the seabed, according to plume modelling. The bulk of the released material is expected to be transported to the seabed within this plume. Some material, however, escapes the plume and becomes suspended in the water column for a longer period. While it was not possible to obtain observations sufficiently close to the release point to observe the initial size distribution exiting the discharge pipe, measurements down-tide of the release indicate that the flocculi (small, strong flocs comprising only a few primary particles) diameter is approximately 15 μ m. Fig. 8 shows depth profiles of the size distributions from the LISST-100x and SilCam close to the release point. These size distributions show median diameters of approximately 20 μ m just below the release (approximately 25 m depth). Below this point, particle sizes increase substantially to a median diameter of approximately 250 μ m. This increase in



Fig. 7. Coloured contour of total volume concentration of particles between 56 and 360 μ m in equivalent circular diameter, as obtained from the SilCam during both ebb and flood transects. The location of the release point is indicated by the yellow square (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)



Fig. 8. Depth profiles of particle size distributions from the SilCam (A) and LISST-100 (B) at STN28 (Fig. 2). The mean size distributions over the depth range of 30–40 m are shown in C.

particle size occurs over a vertical distance of no more than 10 m. Such an increase could be induced by rapid flocculation occurring within the plume, where particle collision frequencies are high. Advection is also likely responsible for exaggerating the apparent effect of flocculation in this case, as the fine particles between 25–32 m depth are those which have separated from the dense plume below. An increase in particle size due to flocculation within plumes such as this has also been reported by other studies of in-situ particle size distribution variability (Mikkelsen and Pejrup, 2000).

While it is difficult to use the present observations to accurately pin down the transport history of the material, the lack of fine



Fig. 9. Montage of randomly-selected particle images from 30–40 m at STN28 (Fig. 2). Particles smaller than 360 μ m are not included.

particles at depth would suggest that they are not transported upwards from the seabed. This is because we observe a shift in d_{50} that is consistent with a shifting modal size in a relatively constant-width log-normal size distribution. If the particles were sourced from an upwards transport mechanism (i.e. from the bottom of the plume or resuspension from the seabed), there would be a vertically-homogeneous concentration of fine material, while coarser material would decrease in concentration away from the seabed.

The mean size distributions from the LISST-100 and the SilCam systems between depths of 30–40 m at STN28, are shown in Fig. 8C. In this case, the value of combined technology for characterising particles is clear, as the LISST-100 shows an explainable variability in the distribution of the fine material that is not resolved by the SilCam, while the high concentrations of larger material at depth is not well resolved by the LISST-100 because the concentration and particle size are approaching the limits of the capability of the instrument.

Large and compact flocs (\sim 300 μ m and upwards) forming in close proximity to the release are not transported far horizontally. These flocs are likely to be high in density with little interstitial water, as can be seen in the montage of particle images selected from the 30–40 m region at STN28 (Fig. 9).

The montages shown in Fig. 9 (and also Fig. 10) are intended as a qualitative illustration of the type and variability of particles within the region represented. They are constructed using a packaging order that iteratively fills the canvas of the figure region, which approximately retain the original particle size distribution (via uniform sampling of the calculated PSD). Therefore it should be noted that the separation of particles in the montages is not necessarily representative of reality—in a high concentration situation the separation between plotted particles will be larger than reality, and vice versa in a low-concentration situation.

A stranded population (Kranck, 1980; Milligan et al., 2007) of small particles are retained within a layer at about 20–25 m depth, and transported horizontally either side of the release point (Fig. 6). The concentration of this fine material decreases with distance from the release, until reaching a non-detectable limit (on the



Fig. 10. Montage of randomly-selected particle images throughout the entire water column from STN02 (Fig. 2). Particles smaller than $360 \,\mu$ m are not included.

OBS) around 3 km west from the release point. These fines likely constitute the smallest particles from the discharge, or the smallest flocculi that cannot be broken any smaller, as there is a sharp separation between the sizes of material advected horizontally and the material sinking immediately below the release.

A remaining question on the fate of the fine material is why the concentration reduces below the detectable limit of the OBS. This may be due to area dispersion and dilution effects, or due to aggregation into larger flocs which settles out of suspension. This question has implications for environmental impact, as low concentration fines can remain in suspension indefinitely and increase the exposure to pelagic organisms, while flocculated sinking material will increase seabed loading and exposure to benthic organisms. The reality is likely a combination of both, the relative proportions of which will be subject to seasonal variability in particle stickiness, 'background' particle concentrations (both biological and inorganic), and hydrodynamic conditions (Engel, 2000; Fettweis and Baeye, 2015).

In addition to the advected fines and high-density flocs, an area containing very long, string-like, low-density flocs (megaflocs) was observed to the west of the release below approximately 25 m (see Fig. 11). In this area some large macroflocs where also seen, but we focus on the string-like megaflocs in the following discussions. While the advected fines can be identified by a 15 μ m peak in the PSD, the low-density flocs are actually similar in median equivalent circular diameter to the high-density flocs close to the release (due to the string-like nature of the megaflocs leading to a small projected area relative to their length). There are two circumstantial aspects supporting the hypothesis that the megaflocs are low in density relative to the similarly-sized flocs close to the release: (1) they are found a substantial distance from the release point and relatively high in the water column, and (2) they appear more complex and string-like in nature when examining particle images from the SilCam (Fig. 10).

Based on the observations of many long, string-like flocs seen in images from the SilCam, it appears that chain-forming diatoms may contribute a significant proportion of suspended organic material occurring naturally within the fjord—at least during the



Fig. 11. Conceptual map showing the dominating particle/floc type in the three different areas discussed in the text.

time of measurement (June). Several studies have investigated aggregation and settling of particulate material relating to diatoms (e.g. Engel, 2000 and references therein). A similar flocculation process between inorganic material and diatom chains has been observed in other coastal sites (Cross et al., 2014). Flocculation between these diatom chains and the advected fine fraction of the mine tailings could therefore act to retain the tailings high in the water column. This type of phytoplankton-mineral flocculation implies that there is potential for significant seasonal variation in floc formation within the region, with a greater presence of the largest, string-like flocs coinciding with a spring diatom bloom. If this is the case, floc size and settling flux could have seasonally variability associated with diatom chain abundances. It is also worth noting that it is likely that such flocs would not be identifiable without particle imaging, as optical scattering measurements will be more sensitive to the smaller, dense components within these flocs, and be relatively insensitive to the diatom (or stringy feature) itself (Graham et al., 2012).

The question of particle orientation may also be of interest, especially in attempting to understand the implication of preferentially-oriented elongated particles in particle collision cross-sections and in optical measurements that are sensitive to the scattering cross-section (Manning, 2004; Milligan, 1995). Elongated particles recorded here were observed in many orientations. Thus, we do not envisage that the imaging volume of the SilCam is contributing to significant alignment of long particles, although instrument-induce turbulence cannot be ruled out in a possible cause for this observation. It may be the case that if there exists a current shear layer, elongated particles may 'fall over' and preferentially orient themselves to align with the streamlines of the flow. This could be an important topic for further study in detailing the responses of multiple measurement techniques in environments which may induce preferentially-oriented elongated particles. Another case which could induce preferential orientations of long particles is an uneven distribution of density along the length of the particles, resulting in one end being less buoyant than the other.

The low-density flocs and high-density flocs have very similar PSD shapes. This implies multiple settling velocities for flocs of equal equivalent diameter. Further work in quantifying the bounds of this range of settling velocities could be usefully applied to modelling the fate and transport of flocculated material. Due to their length, such large, string-like flocs cannot be measured with standard instruments such as the LISST-100 or LISST-Holo, but could contribute significantly to the mass transport and environmental impact inland of the release point. These excessively large flocs appear to have been transported up to two kilometres horizontally from the release, or at least have formed from material that has been transported this far.

It is also not entirely clear why the fine tailings are transported west, but not the low-density large flocs, and why this feature remains persistent during the flood tide (Fig. 6E). This could be related to the time-scale for megafloc formation, where the material that exits the plume is transported west first and is then pumped east, by which time flocs have had opportunity to grow. There could also exist a region of converging flow to the East of the release point, where high diatom abundance converges with the fine mine tailings, stimulating the creation of a new type of aggregate unique to this region. Detailed hydrodynamic and transport modelling may help to test some of these hypothesis, and we plan to pursue this in a future study.

In summary, there are three effective particle populations with different characteristics, which contribute to the total PSD surrounding the discharge location. This conceptual model, which is illustrated in Fig. 11, is discussed in the following section.

3.1. Delineating particle populations

The use of volume (or number) concentrations is commonly adopted for presentation of *in situ* measurements of particle size distributions because optics-based instrument responses are sensitive to particle number and size, and not mass. Using a simple conceptual model (without complex image-based particle classification), relative proportions of volume concentration can be delineated by regions of dense- and low-density flocs, together with the concentration of particles making up the 10–25 μ m peak



Fig. 12. Outline of regions within the fjord cross-section with hypothesised dominance of advected fines (blue), low-density flocs (green), and high-density flocs (red). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

in the PSD. The low-density flocs are dominant within the region to the East of the release below depths of 25 m. The high-density flocs are dominant directly below the release point. An outline of these regions where a specific type of particle dominates is shown in Figs. 12 and 11.

In Fig. 13, size distributions are merged in a similar fashion to that proposed by Mikkelsen et al. (2005), by finding the maximum reported concentration in the overlapping size bins of the LISST-100x and SilCam (excluding the upper four size classes of the LISST-100x), and ignoring particles over $150\,\mu m$ when calculating the concentration of 'Advected Fines'. Such a merging of these size distributions is non-trivial to justify due to differing measurement artefacts between the instruments (Davies et al., 2012; Graham et al., 2012; Davies et al., 2011). However, the smallest four size classes of LISST-100x data are also excluded so as to minimise potential aliasing from out-of-range sizes and particle composition effects (Davies et al., 2012; Andrews et al., 2010). It is important to note that the LISST-100 was only able to capture the size of material in suspension within the far-field where large flocs were low in concentration. The assessment of size distributions that include large flocs is only possible with the inclusion of analysis from the in-situ imaging (in this case, using the SilCam). Routine monitoring of such releases should take this into consideration when determining appropriate instrumentation.

Currently, it is difficult to obtain particle mass estimates together with the size distribution without in situ settling velocity measurements (Friedrichs et al., 2008) or filtration for bulk dry mass. However, it is possible to place some bounds on the mass concentration based on physical limits. Flocs cannot, for example, have a density greater than that of their primary particles, and in this case it would also be reasonable to assume that flocs cannot have a density less than sea water. From these two constraints the mass contributions for each particle type within the fjord can be estimated via scaling of the volume concentrations by assumed densities for each type. Volume distributions for each particle type are shown in Fig. 13. In converting from volume to mass we assume that the density of the advected fines and high-density flocs is equal to the primary particle density $(2.7 \text{ g} \text{ cm}^{-3})$. The lowdensity flocs are more challenging, and so we place bounds on their physically-possible mass distribution. First, the low-density flocs are assumed to have a density of sea water $(1.025 \text{ g} \text{ cm}^{-3})$, producing a mass distribution indicative of the minimum contribution of the low-density flocs to the total mass. The maximum contribution is calculated in the same way, only with the assumption that the low-density flocs have a density equal to the primary particle density of $2.7 \text{ g} \text{ cm}^{-3}$. This results in a range of total relative mass contribution, bound by hard physically-possible limits, of between 2% and 5% from the low-density flocs, and between 85% and 88% for the high-density flocs. The remaining mass would then be contained within the advected fines (7%–13%).

In summary, we propose that the particles that remain in suspension from the tailings discharge (i.e. outside of the primary plume), can be divided into three types, where densely-packaged flocs of approximately 300 µm sink rapidly below the release (likely over the duration of a single tidal cycle), and constitute approximately 85% of the total mass that does not sink inside the primary plume, a fraction of fine material comprising approximately 10% of the release mass is advected higher up in the water column, and the remaining mass is contained within macroflocs and megaflocs that become trapped within the lower water column to the East of the release. We anticipate that this information could be used in conjunction with numerical transport models, where the observed spatially-variant particle size distributions can be used to aid assessments of the significance of flocculation on the transport of the material. Key questions remain, however, on the degree to which the released mine tailings interact with naturallyoccurring particles in the fjord. Further observations are necessary to confirm the significance of organo-mineral aggregation in the area, including seasonally-variant measurements of the large, string-like flocs within the fjord.

4. Conclusions

Measurements of the spatial distribution of suspended material surrounding the submarine tailings placement site in Frænfjorden, Norway, were obtained over flood and ebb transects of the fjord. Data from OBS (turbidity), LISST-100x, LISST-Holo and a bespoke particle imaging system were combined to characterise the size distribution and concentration of suspended particulates of up to 8 mm in diameter.

While useful in determining the distribution of the finest material recorded, OBS and LISST-100x measurements were insufficient at resolving the degree to which large flocs were present within the fjord. The highest concentration of mine tailings were found close



Fig. 13. Delineated volume distributions for the three particle types within the regions shown in Fig. 12: Advected fines (blue), low-density flocs (green), and high-density flocs (red). Dotted lines show the size distribution reported by the LISST-100x, and dashed lines show the size distributions reported by the SilCam. The solid black line in D shows the conceptually-merged size distribution of all particle types. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

to the release point, with some larger complex flocs (several mm in length) transported up to 2 km inland (East of the release). Fines where observed at low turbidity levels up to \sim 3 km west of the release point, but could not be detected beyond this.

We have offered a conceptual model for three dominant classes of particles making up the bulk of our observations outside of the primary plume. Flocculation plays a major role in governing the distribution of material suspended in the water surrounding the discharge. Large, densely-packaged flocs of about $300 \,\mu\text{m}$ in diameter sink rapidly within a relatively small area surrounding the release point. Some fine material does not flocculate immediately, and is observed up to 3 (2) km kilometres west (east) of the release point at a depth of 20-30 m. We also conjecture that this material contributes, after some time, to the formation of megaflocs of several millimetres in length. These very large, lowdensity flocs appear to become trapped to the eastern (inland) side of the release point, and stay resident in the water column for long enough to be transported up to 2 km east of the release. From the long and string-like appearance of these flocs, we suspect that chain-forming diatoms present in the fjord are a catalyst for their formation.

The presented measurements do not permit exact determination of mass concentration from volume concentrations due to lack of information on densities of the observed particles and flocs. However, by combining reasonable assumptions with physical bounds on density, an estimated range of relative mass concentration for different particle classes observed in suspension outside of the primary plume, were obtained: 2%–5% megaflocs and macroflocs, 85%–88% dense flocs and 7%–13% fines.

Acknowledgements

This work was performed within the Research Council of Norway project *Ny Kunnskap Om Sjødeponi* (NYKOS, 236658, 80% financing), co-funded by a consortium of Norwegian Mining Industry Companies (20% financing).

The authors thank Frode Leirvik for his engineering support that enabled deployment of the equipment, and Bror Johansen and Grim Eidnes for their assistance in setting up the current profilers and CTD. Alf, skipper of the boat used ("Emilie"), was extremely helpful and provided first-hand insight into the discharge arrangement. The authors would also like to thank Arnstein Amundsen from Omya Hustadmarmor AS, support from whom was invaluable throughout this study.

References

- Agrawal, Y.C., Pottsmith, H.C., 2000. Instruments for particle size and settling velocity observations in sediment transport. Mar. Geol. 168, 89–114.
- Agrawal, Y.C., Whitmire, A., Mikkelsen, O. a., Pottsmith, H.C., 2008. Light scattering by random shaped particles and consequences on measuring suspended sediments by laser diffraction. J. Geophys. Res. Oceans 113, 1–11.
- Anderson, E.P., Mackas, D.L., 1986. Lethal and sublethal effects of a molybdenum mine tailing on marine zooplankton: Mortality, respiration, feeding and swimming behavior in calanus marshallae, Metridia pacifica and Euphausia pacifica. Mar. Environ. Res. 19, 131–155.
- Andrews, S., Nover, D., Schladow, S.G., 2010. Using laser diffraction data to obtain accurate particle size distributions: the role of particle composition. Limnol. Oceanogr.: Methods 8, 507–526.
- Boss, E., Guidi, L., Richardson, M.J., Stemmann, L., Gardner, W., Bishop, J.K.B., Anderson, R.F., Sherrell, R.M., 2015. Optical techniques for remote and in-situ characterization of particles pertinent to GEOTRACES. Prog. Oceanogr. 133, 43– 54.
- Bowers, D., Hill, P., Braithwaite, K., 2014. The effect of particulate organic content on the remote sensing of marine suspended sediments. Remote Sens. Environ. 144, 172–178.

- Cross, J., Nimmo-Smith, W.A.M., Hosegood, P.J., Torres, R., 2014. The dispersal of phytoplankton populations by enhanced turbulent mixing in a shallow coastal sea. J. Mar. Syst. 136, 55–64.
- Davies, E., Brandvik, P., Leirvik, F., Nepstad, R., 2017. The use of wide-band transmittance imaging to size and classify suspended particulate matter in seawater. Mar. Pollut. Bull. 115, 105–114.
- Davies, E.J., McKee, D., Bowers, D., Graham, G.W., Nimmo-Smith, W.A.M., 2014. Optically significant particle sizes in seawater. Appl. Opt. 53, 1067–1074.
- Davies, E.J., Nimmo-Smith, A.M., Agrawal, Y.C., Souza, A.J., 2011. Scattering signatures of suspended particles: an integrated system for combining digital holography and laser diffraction. Opt. Express 19, 2187–2197.
- Davies, E.J., Nimmo-Smith, W.A.M., Agrawal, Y.C., Souza, A.J., 2012. LISST-100 response to large particles. Mar. Geol. 307-310, 117–122.
- Doe, K., Mroz, R., Tay, K.-L., Burley, J., Teh, S., Chen, S., 2017. Biological effects of gold mine tailings on the intertidal marine environment in Nova Scotia, Canada. Mar. Pollut, Bull. 114, 64–76.
- Dold, B., Bernhard, ., 2014. Submarine tailings disposal (STD) A review. Minerals 4, 642–666.
- Engel, A., 2000. The role of transparent exopolymer particles (TEP) in the increase in apparent particle stickiness (alpha) during the decline of a diatom bloom. J. Plankton Res. 22, 485–497.
- Farkas, J., Altin, D., Hammer, K.M., Hellstrøm, K.C., Booth, A.M., Hansen, B.H., 2017. Characterisation of fine-grained tailings from a marble processing plant and their acute effects on the copepod calanus finmarchicus. Chemosphere 169, 700–708.
- Fettweis, M., Baeye, M., 2015. Seasonal variation in concentration, size, and settling velocity of muddy marine flocs in the benthic boundary layer. J. Geophys. Res. Oceans 120, 5648–5667.
- Friedrichs, C.T., Cartwright, G.M., Dickhudt, P.J., 2008. Quantifying benthic exchange of fine sediment via continuous, noninvasive measurements of settling velocity and bed erodibility. Oceanography 21, 168–172.
- Graham, G., Nimmo-Smith, A., 2010a. The application of holography to the analysis of size and settling velocity of suspended cohesive sediments. Limnol. Oceanogr.: Methods 1–15.
- Graham, G., Nimmo-Smith, W.A.M., 2010b. The application of holography to the analysis of size and settling velocity of suspended cohesive sediments. Limnol. Oceanogr.: Methods 8, 1–15.
- Graham, G.W., Davies, E.J., Nimmo-Smith, W.A.M., Bowers, D.G., Braithwaite, K.M., 2012. Interpreting LISST-100X measurements of particles with complex shape using digital in-line holography. J. Geophys. Res.: Oceans 117, 1–20.
- Hill, P.S., 1998. Controls on floc size in the sea. Oceanography 11, 13-18.
- Hill, P.S., Milligan, T.G., Geyer, W.R., 2000. Controls on effective settling velocity of suspended sediment in the Eel River flood plume. Cont. Shelf Res. 20, 2095– 2111.
- Kach, D.J., Ward, J.E., 2008. The role of marine aggregates in the ingestion of picoplankton-size particles by suspension-feeding molluscs. Mar. Biol. 153, 797–805.

- Kranck, K., 1980. Experiments on the significance of flocculation in the settling of fine-grained sediment in still water. Can. J. Earth Sci. 17, 1517–1526.
- Lyons, M.M., Ward, J.E., Uhlinger, K.R., Gast, R.J., Smolowitz, R., 2005. Lethal marine snow: pathogen of bivalve mollusc concealed in marine aggregates. Limnol. Oceanogr. 50, 1983–1988.
- Manning, A., 2004. Observations of the properties of flocculated cohesive sediment in three western European estuaries. J. Coast. Res. 70–81.
- Mikkelsen, O.A., Hill, P.S., Milligan, T.G., Chant, R.J., 2005. In situ particle size distributions and volume concentrations from a LISST-100 laser particle sizer and a digital floc camera. Cont. Shelf Res. 25, 1959–1978.
- Mikkelsen, O.A., Milligan, T.G., Hill, P.A., Chant, R.J., Jago, C.F., Jones, S.E., Kritsov, V., Mitchelson-Jacob, G., 2008. The influence of schlieren on in situ optical measurements used for particle characterisation. Limnol. Oceanogr.: Methods 6, 133–143.
- Mikkelsen, O.A., Pejrup, M., 2000. In situ particle size spectra and density of particle aggregates in a dredging plume. Mar. Geol. 170, 443–459.
- Milligan, T., 1995. An examination of the settling behaviour of a flocculated suspension. Neth. J. Sea Res. 33, 163–171.
- Milligan, T.G., Hill, P., Law, B., 2007. Flocculation and the loss of sediment from the po river plume. Cont. Shelf Res. 27, 309–321.
- Milligan, T.G., Law, B.A., 2013. Contaminants at the sediment-water interface: implications for environmental impact assessment and effects monitoring. Environ. Sci. Technol. 47, 5828–5834.
- Morello, E.B., Haywood, M.D.E., Brewer, D.T., Apte, S.C., Asmund, G., Kwong, Y. T.J., Dennis, D., 2016. The ecological impacts of submarine tailings placement. In: Hughes, R., Hughes, D.J., Smith, I.P., Dal, A. (Eds.), Oceanography and Marine Biology: An Annual Review, Vol. 54. pp. 315–366.
- Owen, R.B., Zozulya, A. a., 2000. In-line digital holographic sensor for monitoring and characterizing marine particulates. Opt. Eng. 39, 2187.
- Ramirez-Llodra, E., Trannum, H.C., Evenset, A., Levin, L.A., Andersson, M., Finne, T.E., Hilario, A., Flem, B., Christensen, G., Schaanning, M., Vanreusel, A., 2015. Submarine and deep-sea mine tailing placements: A review of current practices, environmental issues, natural analogs and knowledge gaps in Norway and internationally. Mar. Pollut. Bull. 97, 13–35.
- Reynolds, R.A., Stramski, D., Wright, V.M., Wozniak, S.B., 2010. Measurements and characterization of particle size distributions in coastal waters. J. Geophys. Res. 115, 1–19.
- Skei, J.M., 2013. The dilemma of waste management in the mining industry criteria for sea disposal. Mineralproduksjon 3, 1–4.
- Skei, J.M., Syvitski, J.P.M., 2013. Natural flocculation of mineral particles in seawater influence on mine tailings sea disposal and particle dispersal. Mineralproduksjon 3, 1–10.
- Tye, R., Jepsen, R., Lick, W., 1996. Effects of colloids, flocculation, particle size, and organic matter on the adsorption of hexachlorobenzene to sediments. Environ. Toxicol. Chem. 15, 643–651.