



ENVIRONMENTAL MONITORING AT MINE SITES IN GREENLAND

A review of research and monitoring practices and their role in minimising environmental impact

Scientific Report from DCE – Danish Centre for Environment and Energy

No. 364

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Abstract:	This report provides an overview and status of environmental research and monitoring practices near mine sites in Greenland. The most significant mines in Greenland's mining history are described together with research results and monitoring practices used for assessing dispersion, bioaccumulation and toxicological effects of mining pollutants. This knowledge is used to provide recommendations for setting up an adequate environmental monitoring program. Finally, the role of environmental monitoring in regulating mining activities and minimising the associated environmental impact is discussed along with future perspectives and directions for research and monitoring near Greenland mine sites. The report is considered especially relevant for researchers, advisors, mining companies/consultants and people working with the administration of environment and mining in Greenland. However, many of the conclusions and recommendations can also be applied in environmental monitoring and regulation of mining and other activities in other countries, and the report is therefore relevant to a broader audience.
Keywords:	Arctic; Environment; Pollution; Metals; Monitoring programs
Front page photo:	Fieldwork near the Black Angel lead-zinc mine in Maarmorilik, West Greenland, in 2017. The area around Maarmorilik has seen most environmental studies of all Greenland mine sites to date. The Black Angel Mountain is seen in the background and the mine entrances are visible to the left of the black angel-like figure on the mountainside. Photo: Jens Søndergaard.
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Contents

Eqikkaaneq	5
Sammenfatning	7
Summary	8
1 Introduction	9
2 Former and current mine sites in Greenland	11
2.1 The cryolite mine at Ivittuut, South Greenland	13
2.2 The lead-zinc mine at Mestersvig, East Greenland	14
2.3 The Black Angel lead-zinc-silver mine at Maarmorilik, West Greenland	15
2.4 Other mine sites	16
3 Environmental research and monitoring practices at mine sites in Greenland	18
3.1 Overall sampling strategy and targeted pollutants	18
3.2 Measuring dispersion of pollutants in the marine environment	19
3.3 Measuring bioaccumulation in the marine environment	21
3.4 Measuring dispersion and bioaccumulation of pollutants in the terrestrial and freshwater environment	23
3.5 Measuring or assessing toxicological effects of pollutants	26
4 Considerations for setting up an environmental monitoring program at Greenland (and other Arctic) mine sites	29
5 The role of environmental research and monitoring in minimising environmental impact of mining in Greenland	31
6 Future perspectives	33
7 Final remarks	34
8 References	35
9 Appendices	41
Appendix 1. Methods for sample preparation and chemical analyses	41
Appendix 2. Practices for storage of samples and data	44

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Eqikkaaneq

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Sammenfatning

Denne rapport giver en oversigt og status for miljøforskning og praksis til miljømonitoring ved grønlandske miner. Ligeledes beskrives, hvordan resultater fra forskningen og monitoringen er blevet brugt til miljøregulering og minimering af miljøeffekter. I rapporten beskrives de vigtigste miner i Grønlands historie sammen med de vigtigste forskningsresultater og metoder til bestemmelse af spredning, biologisk akkumulering og toksiske effekter af forurenende stoffer. Efterfølgende gives en række anbefalinger til etablering af et miljømonitoringsprogram, og endelig diskuteres fremtidige perspektiver og retninger for forskning og monitoring ved grønlandske miner.

Rapporten er især relevant for forskere, rådgivere, mineselskaber og deres konsulenter samt embedspersoner, der arbejder med miljø og minedrift i Grønland. Imidlertid kan mange af konklusionerne og anbefalingerne også overføres til miljømonitoring og miljøregulering ved miner og andre industrielle aktiviteter andre steder i verden, hvorfor målgruppen er væsentlig bredere.

Minedrift begyndte for alvor i Grønland i 1850'erne med åbning af kryolitminen ved Ivittuut i Sydgrønland. Siden da er minedrift efter bly, zink, sølv, guld, olivin, rubiner og feldspat (og i mindre grad kul, grafit og kobber) foregået ved et antal miner. Minedrift ved tre af de tidligere miner, kryolitminen ved Ivittuut, bly-zinkminen ved Mestersvig og bly-zinkminen ved Maarmorilik, medførte betydelig forurening (primært med bly og zink) i det omkringliggende miljø. Dette var især pga. deponering i miljøet af restprodukterne fra minedriften, såkaldt gråbjerg og tailings. Disse tidligere mineområder har efterfølgende været anvendt som vigtige studieområder for udvikling af metoder til miljømonitoring i Grønland.

Resultater fra forskningen viser, at miljømonitoring ved grønlandske miner foretages mest hensigtsmæssigt ved måling af både ikke-biologiske prøver (vand, sediment, jord og støv) og biologiske prøver af udvalgte monitoringsorganismer. Måling af udledninger og emissioner af forurenende stoffer skal tage hensyn til de tidsmæssige variationer, der forekommer i Arktis (som f.eks. den typiske høje udvaskning om foråret knyttet til snesmeltning mv.). Udvælgelsen af monitoringsorganismer afhænger af faktorer såsom tilstedeværelse, specifikt hvilke forurenende stoffer der er relevante, og hvordan stofferne forekommer i miljøet, men blåmuslinger, blæretang, ulke, fjeldørreder og lav er typisk anvendte organismer til miljømonitoring ved miner i Grønland.

Resultaterne fra forskningen og miljømonitoringen er blevet anvendt til løbende vurdering af miljømæssige effekter ved igangværende og nedlukkede miner og i nogle tilfælde også til regulering af igangværende mineaktiviteter med henblik på at minimere den miljømæssige påvirkning. Resultaterne er også blevet anvendt i forbindelse med fastsættelse af grænseværdier til udledninger indskrevet i tilladelserne.

Den fremtidige miljømonitoring ved grønlandske miner vil inkludere en række yderligere værktøjer til vurdering af miljømæssige effekter i takt med en fortsat forskning i disse områder og teknologiske fremskridt. Den fremtidige monitoring kan også inkludere monitoring på regionalt niveau for at undersøge kumulative effekter og effekter af aktiviteter relateret til minedrift i større skala.

Summary

This report provides an overview and status of environmental research and monitoring practices near mine sites in Greenland and how the results of research and monitoring have been applied to regulate mining activities and minimise the associated environmental impact. The most significant mines in Greenland's mining history are described together with research results and monitoring practices for assessing dispersion, bioaccumulation and toxicological effects of pollutants. Finally, considerations for setting up an adequate initial monitoring program are given together with future perspectives and directions for research and monitoring at Greenland mine sites.

The report is especially relevant for researchers, advisors, mining companies/consultants and people working with the administration of environment and mining in Greenland. However, many of the conclusions and recommendations can also be applied to environmental monitoring and regulation of mining and other activities in other countries, and the report are therefore relevant to a broader audience.

Mining in Greenland really began in the 1850s with the opening of the cryolite mine in Ivittuut in South Greenland. Since then, mining for lead, zinc, silver, gold, olivine, rubies and feldspar (and to a small extent coal, graphite and copper) has taken place at a number of mine sites. Mining activities at three former mine sites, the cryolite mine in Ivittuut, the lead-zinc mine in Mestersvig and the lead-zinc mine in Maarmorilik, caused significant metal pollution (mostly with lead and zinc) in the surrounding environment. This was mainly due to the disposal of mine waste (i.e. waste rock and tailings) into the environment. These polluted former mine sites have later served as field laboratory study areas for the development of methods for pollutants monitoring.

Results show that environmental monitoring at Greenland mine sites is most adequately conducted using a combination of non-biota (water, sediment, soil and dust) and biota (i.e. biological monitoring organisms). We highlight the need to take temporal variations unique to the Arctic into account with respect to the discharge of pollutants (such as the typical flush of pollutants during spring associated with snowmelt). Biological monitoring organisms need to be selected from a diverse suite depending on abundance, pollutants of concern, speciation of pollutants etc. Typically, blue mussels, seaweed, sculpins, lichens and Arctic char are used as key monitoring organisms.

Results from environmental research and monitoring programs in Greenland have been used to establish environmentally safe threshold levels for regulation of mining activities during the permitting stage and for continuous evaluation, and in some cases also regulation, of ongoing activities in order to minimise environmental impact.

Future environmental monitoring of mining activities in Greenland will include a range of additional tools for assessing environmental impact owing to continuation of applied research in mining areas and technological advances. Future monitoring programs may also include regional monitoring programs to investigate cumulative effects of mining activities on a larger scale.

1 Introduction

Environmental pollution from mining activities is a well-known risk and concern, which in the western world in recent decades has been addressed by comprehensive regulation (e.g. EU, 2006; EC 2009). As a supplement to regulation of mining activities during the permitting stage, environmental monitoring during the construction, operation and post-mining phases can be used to assess compliance with and identify pollution from ongoing or past activities. Based on the monitoring results, further regulation or mitigation actions may be introduced to reduce the pollution and impacts from mining. In a long-term perspective, monitoring results from past mining operations can also often provide valuable knowledge that can be implemented in regulations of new mine sites to minimise environmental impact.

Greenland has a long history of mining, starting with industrial extraction of cryolite in 1854 in South Greenland. The first environmental studies at Greenland mines began in the early 1970s following the opening of the lead-zinc mine in Maarmorilik in West Greenland (Asmund et al., 1994). Like in the rest of the world, some of the old Greenland mines have a legacy of long-lasting pollution. However, research and monitoring at these polluted sites have enabled studies on the dispersion, bioaccumulation and effects of mining pollution under arctic conditions and provided information for the development of a regulatory system to minimise the impact of mining activities.

Since the early 1970s, monitoring of Greenland mine sites has been performed regularly at both operating and closed mines. Danish Centre for Environment and Energy (DCE) (formerly named National Environmental Research Institute) and before that Greenland Environmental Research Institute and Greenland Fisheries and Environmental Research Institute has conducted environmental monitoring at Greenland mine sites for the Greenland authorities since the very beginning. In the last decade, the environmental monitoring has been done in collaboration with the Greenland Institute for Natural Resources (GINR). DCE and GINR are advisors to the Greenland authorities on all environmental issues related to mining.

DCE and GINR hold/administer a large sample- and databank for the Greenland authorities. It contains environmental samples and data from former and present mine sites that are used in the regulation of mining activities and for research purposes. In addition, DCE has an ISO 17025 accredited trace metal laboratory and a radioecology laboratory used for chemical and radiological analyses of the environmental samples. Historically, monitoring at operating mine sites has been performed by the mining companies (high-frequency water and tailings sampling at outlets etc.) combined with audit monitoring by DCE and GINR on behalf of the Greenland authorities. The audit monitoring has typically been based on a wide range of annual samples taken in the mining area and in adjacent areas. Monitoring after mine closure has been conducted by DCE and GINR on behalf of the authorities.

An important foundation for evaluating environmental changes at operating or former mine sites is knowledge about the environmental conditions prior to the activities (so-called baseline conditions). This is important because elevated concentrations of potential pollutants (e.g. metals) contained in the ore or waste rock often occur naturally at the mine sites. According to the current

practice, two-three years of baseline data are needed (MRA, 2015). In the past, DCE conducted the baseline studies at Greenland mine sites as part of their Environmental Impact Assessment (EIA) work, but in recent years, the task has been undertaken by the mining companies and their consultants. All mining companies operating in Greenland are obliged to draw up an EIA before being granted a license for exploitation. The EIA must give a correct and fulfilling assessment of the environmental impact from the activity. DCE and GINR have advised on the development of specific guidelines for the EIA work including environmental programs (MRA, 2015) and a set of water and air quality criteria for Greenland mining activities. DCE and GINR are subsequently involved in the assessment of the EIA for the authorities. A correct and fulfilling EIA is one of the documents needed before the Greenland Self-Government can decide for or against a specific mining project.

The methods and techniques for environmental monitoring have evolved over the years due to technological advances and experience gained from previous monitoring and research. DCE and GINR aim to continue research related to the environmental monitoring of Greenland mine sites to improve the methods for assessing the environmental impacts of mining as well as to gain more knowledge of these.

This review aims to provide an overview of environmental research and monitoring practices at mine sites in Greenland and draw conclusions from the experience gained so far. The most significant mines in Greenland mining history are described, and studies and methods for assessing the dispersion, bioaccumulation and toxicological effects of pollutants from mining are presented. Finally, the role of environmental research and monitoring to minimise environmental impact in Greenland is highlighted with examples and future directions and perspectives for environmental monitoring at Greenland mine sites are given.

2 Former and current mine sites in Greenland

The location of former and present mine sites in Greenland is shown in Figure 1 and more detailed information on the mines is provided in Table 1. Greenland is extraordinary as it consists of a large ice-free land area of about 410,000 km², spanning over more than 20 degrees latitude. Greenland is very thinly populated with about 56,000 inhabitants and no roads to connect the cities/towns/settlements. It is a general feature of mines in Greenland that they are located close to the sea or fjords to minimise costs as there is hardly any road infrastructure outside the cities/towns/settlements.

Figure 1. Map of Greenland with major former and present mine sites.



Table 1. Major former and present mine sites in Greenland with some environmental characteristics.

Mine	Ore type	Mining period	EIA	First environmental monitoring/study	Major environmental issue(s)	Environmental legacy
Ivittuut	Cryolite	1854-1987	No	1982	Pollution with mainly lead and zinc from waste rock situated in the tidal zone between the mine pit and the Arsuk Fjord. Transport of lead and zinc from the pit water to the fjord.	The Arsuk Fjord and a 3-4 km area outside it were still polluted with lead and zinc during the last study in 2013. A decreasing pollution trend has occurred since the first environmental study in 1982.
Qullissat	Coal	1924-1972	No	2015	None ¹	None ¹
Mestersvig	Lead-zinc	1956-1963	No	1979	Pollution with mainly lead and zinc from: 1) tailings disposed of in Tunnel River; 2) dust dispersion of concentrate along the haul road; 3) spills of concentrate at Nyhavn (harbour).	The area was still polluted with mainly lead and zinc near the mine, along the haul road, at Nyhavn and in the adjacent fjord during the last study in 2014. A general trend toward decreasing pollution has been observed since 1991.
Maarmorilik	Lead-zinc-silver	1973-1990	No	1972	Pollution with mainly lead and zinc from: 1) tailings discharged into the Affarlikassaa Fjord; 2) waste rock deposited on the mountain slopes and in the fjords; 3) residues of ore and concentrate from the mining town.	The Qaamarujuk Fjord was still polluted with mainly lead and zinc during the last study in 2017. Elevated concentrations were observed in mussels and seaweed c. 12 km from the mine. Dust dispersion of mainly lead and zinc was still significant, presumably derived mainly from degrading waste rock at the mountain slopes. The pollution decreased immediately upon the mine closure in 1990, but a significant trend toward decreasing pollution in the area has not appeared during the past c. 20 years.
Nalunaq	Gold	2004-2013	Yes	1998	None. Cyanide was used in the production, but no significant dispersion of cyanide was observed in the monitoring.	None.

Seqi	Olivine	2007-2009	Yes	2004	Dust dispersion during the mining period, measurable 5-7 km from the mine with elevated concentrations of chromium, nickel, iron and cobalt in lichens.	None. In the last monitoring in 2018, concentrations had decreased to a level where elevated concentrations in surface soil and lichens due to dust dispersion/deposition were only measured locally (within c. 1 km of the mine).
Aappaluttoq	Rubies	2017-	Yes	2006	None	None
White Mountain	Anorthosite	2018-	Yes	2012	None	None

¹The first environmental screening study conducted in 2015 did not identify any significant chemical pollution of the surrounding environment at Qullissat, either from the Qullissat mine site or from the town of Qullissat. However, it does not rule out past pollution.

2.1 The cryolite mine at Ivittuut, South Greenland

The cryolite mine at Ivittuut in the Arsuk Fjord in Southwest Greenland started operating in 1854 and the production lasted for more than 130 years until the mine was finally closed in 1987. Cryolite (with the chemical formula Na_3AlF_6) is a very rare mineral and the mine at Ivittuut was the only major cryolite mine worldwide. Cryolite is an industrial mineral that is used as a flux agent in the electrolytic process of aluminium extraction from the aluminium-rich oxide ore bauxite. Today, due to the scarcity of natural cryolite, synthetic cryolite is produced from the mineral fluorite. The production at Ivittuut peaked in 1943 with 80,000 tonnes cryolite, and during the entire mining period a total of 3.7 million tonnes cryolite were produced (Johansen et al., 2010b). The ore was blasted, crushed and sorted on site and shipped to Denmark for further processing. The mine was an open-pit mine situated in immediate vicinity to the shoreline and waste rock was used as landfill between the pit and the fjord. The landfill was open to the tidal movements in the fjord, saturating the rocks with seawater.

The first environmental studies conducted in the area in 1982 revealed significant pollution with mainly lead and zinc in the Arsuk Fjord due to the mining activity. The pollution was mainly related to the transport of lead and zinc from the waste rock (present as the sulphide minerals galena and sphalerite, respectively) situated at the coastline. In 1985, it was estimated that between 400 and 1,000 kg dissolved lead entered the Arsuk Fjord from the waste rock. This transport has decreased since then (Johansen et al., 2010b; Bach et al., 2014a). The first studies showed that the species affected by the heavy metal pollution were brown seaweed (*Fucus vesiculosus*) and blue mussels (*Mytilus edulis*), while fish and prawns from the fjord were not impacted. Consequently, the environmental studies, carried out later at Ivittuut annually in the 1980s and every two-three years during 1990-2013 had focus on these two species (Bach et al., 2014a).



Photo 1. The abandoned mine pit and remaining buildings at the former cryolite mine in Ivittuut in 2015. Photo: Morten Birch Larsen.

2.2 The lead-zinc mine at Mestersvig, East Greenland

The lead-zinc mine at Mestersvig in East Greenland was in operation for a short period between 1956 and 1963. The mine was underground and located c. 10 km inland from Kong Oscar Fjord. During the mining period, a total of 554,000 tonnes ore were produced, yielding 58,000 tonnes lead concentrate and 75,000 tonnes zinc concentrate. Lead and zinc were contained as sulphide minerals (galena and sphalerite, respectively) in the ore. The ore was processed on site and tailings from the ore-processing were deposited on a mountain slope near the mine. The lead/zinc concentrate was transported in bags from the mine to the small harbour of Nyhavn where it was stored until loading and shipment.

The first environmental studies were conducted in 1979 and the results revealed significant pollution with lead and zinc in the area, both in the terrestrial and the marine environment. Subsequent environmental studies were undertaken in 1983, 1985, 1986, 1991, 1996, 2001 and 2014. The two main sources of pollution were considered to be the remains of the tailings dispersed both as dust and via the river Tunnel River and spills of concentrate off the quay in Nyhavn. Elevated levels of lead and zinc were observed in water, soil and sediment as well as in lichens, vascular plants, seaweed, three species of bivalves and sculpins (Johansen et al., 2008; Aastrup et al., 2017). Seals in Kong Oscars Fjord did not show elevated heavy metal levels compared with elsewhere in Greenland (Johansen et al., 2008).

Photo 2. Nyhavn at Mestersvig in 2014. The mine is located c. 10 km inland. During the mining period (1956-63), lead and zinc concentrate was shipped out from Nyhavn and elevated concentrations can still be measured (in 2014) in both the terrestrial and marine environment near Nyhavn. Photo: Lis Bach.



2.3 The Black Angel lead-zinc-silver mine at Maarmorilik, West Greenland

The Black Angel mine at Maarmorilik in West Greenland operated from 1973 to 1990. The mine was underground with the mine entrances – located at 600 m altitude on the Black Angel Mountain – that were connected to a mining town at sea level by cable car. The ore consisted of lead (4%), zinc (12%) and silver (30 ppm) contained in the sulphide minerals galena and sphalerite. The remaining ore contained marble and pyrite. The ore was transported to the mining town where it was processed into concentrate, stored and shipped. The total production during 1973-90 was 11.2 million tonnes ore, resulting in 590,000 tonnes lead concentrate (70% lead, 420 ppm silver) and 2,327,000 tonnes zinc concentrate (58% zinc) (Thomassen, 2003).

The mountainous topography prevented the design of a land-based tailings disposal system, and the tailings from the ore processing were therefore discharged into the small fjord Affarlikassaa (named the A-fjord in the following) that is partly separated from the outer fjord Qaamarujuk (named the Q-fjord) by a sill at the fjord mouth (Asmund et al., 1991). Waste rock from excavation of the mine tunnels, which contained elevated concentrations of lead and zinc (c. 1% lead and 3% zinc), was deposited on the mountain slopes. The largest of the waste rock dumps (the North Face Dump), which comprised roughly 400,000 tonnes of rock, extended down into the fjord. About 90% of the North Face Dump was removed as part of the mine closure in 1990 and deposited on top of the tailings in the A-fjord (Asmund, 1992a).

The first environmental studies were initiated in the early 1970s and studies were continued in the area at 1-3 year intervals until 2012, followed by a study in 2017. Significant pollution with metals, mainly lead and zinc and to a lesser extent cadmium, silver, arsenic and mercury, was observed in the Maarmorilik area because of the mining activity. Elevated concentrations of metals were measured in seawater, sediment and a number of species in both the terrestrial and marine environment (Larsen et al., 2001; Elberling et al., 2002; Johansen et al., 2010a; Søndergaard et al., 2011a and b; Sonne et al., 2014). During the mining period, both the tailings and the waste rock acted as significant sources of pollution. Disposal of tailings into the A-fjord turned out to be a

major source of pollution to the area due to a combination of metals dissolving from the tailings and seasonal transport of pollutants from the A-fjord to the Q-fjord as a result of complex hydrographic processes (Asmund, 1992b; Poling and Ellis, 1995). The key hydrological processes for dispersion of pollutants from the A-fjord were seasonal vertical mixing of the water layers in the A-fjord and complete flushing of pollutants from the A-fjord during some winters due to in- and outgoing currents (Møller, 1978, Lewis and Perkin, 1982; Møller et al., 1982; Møller and Pedersen, 1983).

After the mine closure, pollution from the tailings and the waste rock deposited in the A-fjord decreased markedly due to burial of the tailings and waste rock caused by natural sedimentation processes. In recent years, remains of waste rock on the mountain slopes and in the coastal area, especially around the remains of the North Face Dump, together with residues at the mining town are considered to be the main sources of pollution (Søndergaard et al., 2011a). Due to the pollution status of the area and the relatively easy access to the site, Maarmorilik has been subject to numerous studies related to environmental assessment and method development, and it is by far the most thoroughly studied mining area in Greenland.

Photo 3. The former Black Angel lead-zinc mine in Maarmorilik, West Greenland, in 2017. During the mining period (1973-1990), a cable car connected the mining town and the ore processing facility situated in the foreground to the mine entrances located at c. 600 m altitude on the steep mountainside seen in the background. Photo: Jens Søndergaard.



2.4 Other mine sites

The cryolite mine at Ivittuut and the lead-zinc mines at Mestersvig and Maarmorilik are the most important of the former mines in Greenland from an environmental perspective. At these sites, tailings and waste rock were identified as significant sources of pollution in the surrounding area, which has induced frequent environmental monitoring and numerous studies and assessments. There have been other mines in the Greenland mining history, as briefly described below, but no significant pollution from these sites to the surrounding environment has been observed (although the old (pre-1970s) mine sites were not monitored during operation).

The mining history of Greenland started in 1782 with small-scale coal mining at the Disko Island in West Greenland (Secher and Sørensen, 2014). Later, during the nineteenth century, a number of other small-scale (and short-lived) mines were opened for exploitation of coal, copper, graphite and marble.

Apart from the mines at Ivittuut and Mestersvig, the most significant of the earlier mines was a coal mine at Qullissat on the Disko Island (Figure 1), which operated from 1924 to 1972 and produced 570,000 tonnes coal. A recent study evaluated the environment at Qullissat and found no indication of significant chemical pollution at the site (Søndergaard et al., 2017).

In recent years, there have been four active mines in Greenland: 1) a gold mine in Nalunaq in South Greenland, which operated from 2004 to 2013; 2) an olivine mine at Seqi in Southwest Greenland, which operated from 2007 to 2009; 3) a ruby mine at Aappaluttoq in Southwest Greenland, which opened in 2017 and is currently in operation; 4) a feldspar (anorthosite) mine at 'White Mountain' in Southwest Greenland, which opened in 2018. None of these mines have had any significant environmental impact (Bach and Larsen, 2016; Søndergaard, 2019), likely thanks to strict environmental regulation and frequent and comprehensive environmental monitoring and control.

3 Environmental research and monitoring practices at mine sites in Greenland

3.1 Overall sampling strategy and targeted pollutants

Most environmental studies at Greenland mine sites are based on the collection and analyses of samples from a range of sampling stations established in the fjords, along the coastline and inland (typically 25-50 stations in total). Most sampling stations are typically established near the potential sources of pollution to most accurately assess pollution levels and identify sources, while the remaining sampling sites are located in a gradient away from the mine. The dominant directions for dispersion by wind or currents are also taken into account, placing more stations in the dominant directions. If fresh water samples are taken in streams or lakes, sampling stations are typically placed both upstream and downstream from the potential pollution source and at the dominant inlets and outlets of the lakes.

Seasonal variations in water chemistry can be very pronounced in Greenland, for example due to spring flushes of winter-accumulated pollutants on land released during thaw and snowmelt (Søndergaard et al., 2012). The sampling period for freshwater are therefore set to cover the expected variability. In most studies, reference samples from one or two sampling stations far away from the mine (and therefore unlikely to be influenced by the mining activities) are used for comparison. For more recent mines, where environmental baseline studies have been made prior to the actual mining, sampling stations are typically reused in further monitoring studies because it allows the most accurate comparison of sample composition before and after mining. Finally, accessibility is an important parameter and sampling stations are typically placed to allow access by foot or boat to avoid the high cost of helicopter transportation.

As for the targeted pollutants in the environmental studies, focus is typically directed at metals and other elements found to be elevated in the ore and mine waste products compared with natural rocks or sediments in the area or in the Earth's crust. Today, modern analytical methods like Inductively Coupled Plasma Mass Spectrometry (ICP-MS) enable analyses for most of the elements in the periodic table (60+ elements) in a single scan and such analyses therefore provide a very solid and comprehensive basis for targeting potential pollutants. Besides metals and other (non-radioactive) elements, other potential pollutants include radionuclides, oil and chemicals or additives used in ore-processing at the mine site.

At the former mine sites in Greenland, the main targeted pollutants is lead and zinc (Maarmorilik, Ivittuut and Mestersvig), cadmium, silver, arsenic and mercury (Maarmorilik) and chromium and nickel (Seqi). At the former gold mine in Nalunaq, cyanide used in the extraction process was monitored frequently besides elements such as copper, chromium, arsenic and cobalt, which were found in the ore together with the gold. In connection with the recently proposed mines at the alkaline syenite-type deposits in Kvanefjeld and Kringlerne in South Greenland, potential pollutants of concern will also include fluoride, rare earth elements and radionuclides (uranium, thorium and their decay series products).

Methods for sample preparation and chemical analyses and practices for storage of samples and data are described in detail in Appendix 1-2.

3.2 Measuring dispersion of pollutants in the marine environment

Since the first environmental studies at the Black Angel mine in the 1970s and 1980s, dispersion of pollutants in the marine environment in Greenland has been assessed using seawater samples and samples of bottom sediment in the fjords (Bondam, 1978; Loring and Asmund, 1989).

Seawater samples have typically been collected at regular depth intervals (approximately every 5 or 10 m) from surface to bottom using metal-free water samplers (like the Hydro-Bios standard or reversible water samplers) or pumped up through a polyethylene tube (Johansen et al., 2006). Subsequently, seawater samples have been filtered on-site through 0.45 µm size filters to obtain samples of dissolved pollutants in the water. In some studies, also the suspended particulate matter on the filters has been quantified and analysed to obtain the concentration of suspended particulate-bound pollutants in the water (Loring and Asmund, 1989; Søndergaard et al., 2011a). To gather temporal information on the seawater composition, sampling of seawater has sometimes been performed both during summer and winter (Loring and Asmund, 1989). This was the case at the Black Angel mine in the 1970s and 1980s since a dispersion of pollutants was observed that could not be explained by the summertime seawater measurements.

In recent years, passive chemical samplers (i.e. Diffusive Gradients in Thin films, DGT) that can measure dissolved labile metal species in both sea- and freshwater have been used in addition to conventional techniques for measuring dissolved metals in seawater (Søndergaard et al., 2014). DGT samplers have the advantage that they provide a measure of the time-integrated and 'labile' metal concentrations during the deployment period, which can be up to several weeks, as opposed to conventional water sampling that only provides a snapshot of the water chemistry. DGT samplers are also relatively easy to handle and analyse (especially compared with seawater samples). A technique to keep DGTs suspended in the water column is a buoy-setup with an anchor to the seafloor and an adapter to mount the samplers on. DGT samplers, however, only work for some metals depending on the type of the sampler. Also, water quality criteria are typically only established for total metal concentrations in sea- and freshwater (MRA, 2015), which requires conventional water sampling. Consequently, DGT samplers should be regarded as a supplement to rather than a substitute for conventional water sampling.

Bottom sediment samples taken either as grab or core samples have been used to evaluate the dispersion, sedimentation rate and impact of pollutants on the seafloor (Loring and Asmund, 1989; Elberling et al., 2002; Perner et al., 2009; Søndergaard et al., 2011a). Grab samplers (e.g. Van Veen- or Ekman-type sediment grabs) are appropriate for collecting the upper few centimetre layers of sediment and have the advantage that they are relatively light and easy to handle manually in a small boat. Core samplers enable collection of sediment cores up to approximately 1 m, but they are heavy and require a larger ship and a winch. At the Black Angel mine in Maarmorilik, Loring and Asmund (1989) used layers of a core sample using ^{210}Pb (lead-210) radiometric dating to assess the sedimentation rate of polluted sediment. Elberling et al. (2002) took this a step further and used a series of ^{210}Pb -dated sediment cores to estimate the size

of the dispersion area and the accumulation rate of lead and zinc in bottom sediment within the same area during a 100 year-period. Furthermore, Elberling et al. (2002) used ratios between pollutants (lead/zinc) and stable isotopic signatures of lead (i.e. ^{206}Pb , ^{207}Pb and ^{208}Pb) in bottom sediment to study mobilisation and transport processes and to identify lead from the mining deposit versus natural background lead in the area. Stable isotopic signatures of lead were later used to identify lead from the mining deposit versus natural background lead in biota (Søndergaard et al., 2010). A difference in the grain size of sediment between sites or within a core can bias a direct comparison between concentrations of pollutants since smaller particles have a larger specific surface area and the ability to adsorb pollutants (Quevauviller et al., 2011). To normalise for differences in grain size of the sediment within the cores or between cores or grab samples, the ratio between the pollutant and aluminium (regarded as a conservative tracer) has been used in a number of studies to improve the basis for comparison (Perner et al., 2009, Søndergaard et al., 2011a). In other studies, sediment has been sieved to a certain fraction (e.g. <1 mm or <0.063 mm) and only the chemical composition in that size fraction was reported (Asmund et al., 1991; Søndergaard et al., 2019).

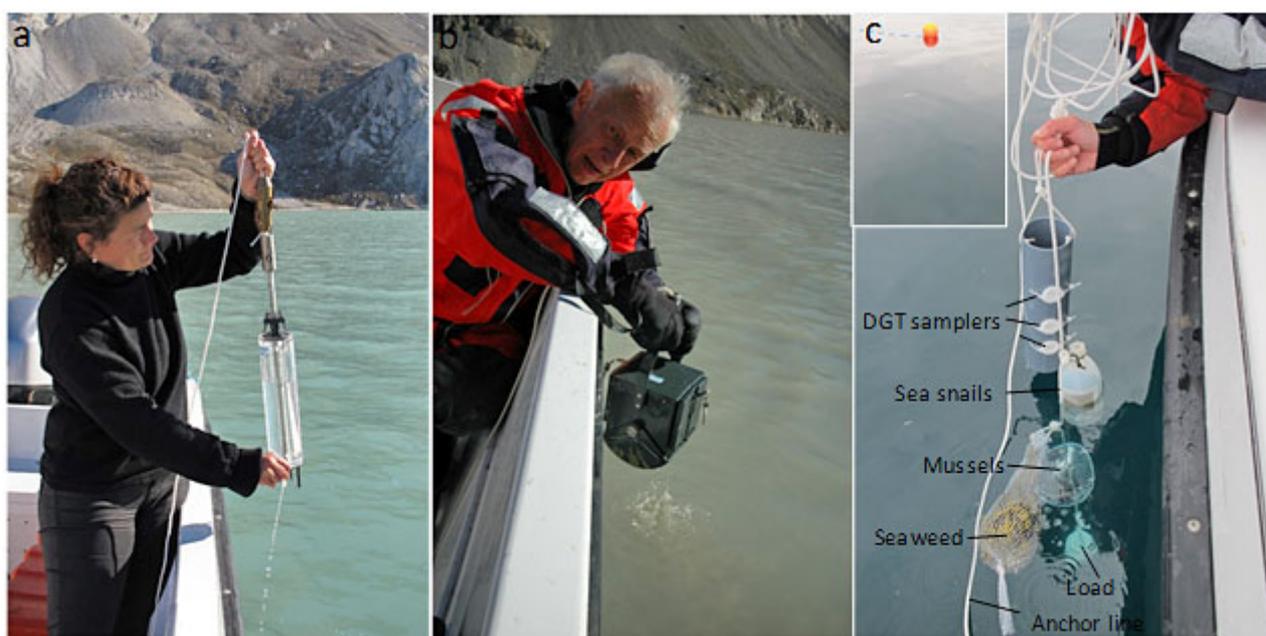


Photo 4. Dispersion of pollutants in the marine environment is typically measured using: (a) depth-specific seawater samples, (b) grab samples of surface sediment, (c) DGT passive chemical samplers left suspended in the water column. DGT samplers enable depth-specific time-integrated measurements (from typically days to weeks) of dissolved labile metals. DGT samplers are deployed from buoys anchored to the seafloor. In addition to DGT samplers, deployment of transplanted sea snails, blue mussels and seaweed has been used successfully to assess depth-specific short-term biological uptake of pollutants (Søndergaard et al., 2014). All photos: Jens Søndergaard.

In addition to sampling and measurements of water and sediment, oceanographic studies have been performed to understand currents, mixing and dispersion of water masses and pollutants in the marine environment. During such studies near the Black Angel mine, Møller (1984) found that seasonal hydrographical processes were responsible for dispersion of pollutants from marine-deposited tailings into a much larger area than originally anticipated. Tailings were deposited at c. 30 metres depth in the small A-fjord that was separated from the larger Q-fjord system by a sill at c. 20 m depth. During summer, the saline-polluted bottom water in A-fjord was stagnant and sepa-

rated from the upper water layer by a halocline. During some winters, however, formation of sea ice and the lack of freshwater input caused formation of a saline surface layer that was substantial enough to sink down and create vertical mixing of the water in the fjord. Furthermore, during those winters (four out of seven winters during the period 1977-84), water entered A-fjord from the outer fjord as a bottom current replacing the existing bottom water, which led to complete flushing of suspended or dissolved pollutants from A-fjord to the outer fjord system.

3.3 Measuring bioaccumulation in the marine environment

Since the first environmental studies at Maarmorilik in the 1970s, seaweed (*Fucus spp.*), blue mussels (*Mytilus spp.*) and sculpins (*Myoxocephalus spp.*) have been used as key monitoring organisms for measuring bioaccumulation in the marine environment near mine sites in Greenland (Johansen et al., 1991; Søndergaard et al., 2011a). In addition, bioaccumulation in prawns (*Pandalus borealis*), sea snails (*Littorina saxatilis*), amphipods (*Gammarus spp.*), other mussels like *Mya truncata*, *Macoma calcarea* and *Musculus discors* and other fish species like Greenland cod (*Gadus ogac*), Atlantic cod (*Gadus morhua*), capelin (*Mallotus villosus*), Greenland halibut (*Reinhardtius hippoglossoides*) and spotted wolf fish (*Anarhichas minor*) have been studied (Johansen et al., 1991; Larsen et al., 2001; Søndergaard et al., 2014). A recent study also evaluated the potential use of green sea urchins (*Strongylocentrotus droebachiensis*) as a monitoring organism (Søndergaard et al., 2019). The following describes the use of seaweed, blue mussels and sculpins as they are by far the most studied and applied organisms for bioaccumulation monitoring at Greenland mine sites. This is because these organisms: 1) are abundant at most mine sites in Greenland, 2) are stationary/sedentary, 3) effectively accumulate pollutants such as metals and 4) represent different trophic levels/habitats (i.e. have different food sources). Using monitoring organisms from different trophic levels/habitats allow detection and assessment of, for instance, time-integrated heavy metal loading from a broad range of pollution sources.

Seaweed (*Fucus vesiculosus* or *Fucus distichus*) collected from the tidal zone has been part of the environmental monitoring programs at the former mine sites in Maarmorilik, Mestersvig, Ivittuut, Seqi and Nalunaq (Johansen et al., 2008; Johansen et al., 2010a and b; Søndergaard, 2013a; Bach and Larsen, 2016). After collection, the green-coloured growth tips of the seaweed are cut off and sampled for subsequent chemical analyses. Accumulation of metals in seaweed is regarded as a relative measure of the dissolved metal concentrations within the seawater (Rainbow, 1995) and growth tip metal concentrations reflect the accumulation in the present growing season (spring-autumn) as indicated in previous studies of seaweed metal accumulation (Søndergaard et al., 2011a).

Blue mussels (*Mytilus spp.*) collected from the tidal zone have been part of all the environmental monitoring programs mentioned above, except from Mestersvig on the east coast of Greenland where blue mussels are absent. Recent studies have shown that two species of blue mussels occur in West Greenland, *Mytilus edulis* and *Mytilus trossulus* (Wenne et al., 2016; Bach et al., 2018). Blue mussels are suspension feeders that filter large volumes of water through their gills (typically c. 3 litres per hour for an adult mussel) and feed mainly on phytoplankton (Beyer et al., 2017). Consequently, metal accumulation in mussels is considered a relative measure of both dissolved and particle-bound metals in the seawater (Rainbow, 1995). Typically, the mussels are divided into 1 cm size groups (4-5 cm, 5-6 cm etc.), cut open and left to drain for a

couple of minutes. Subsequently, all soft parts are cut out and sampled. A sample size of 20 individuals pooled into one sample is preferred for each size group. Also, the precise mean shell length (to the nearest 0.1 cm) of the mussel shells in each pooled sample is determined. Since metal concentrations may depend on the size of the mussels (Rigét et al., 1996), a comparison between mussels of the same size is preferable.

Another aspect to consider is the slow and incomplete excretion of pollutants by mussels in case the pollution decreases. This has been shown by Rigét et al. (1997) who transplanted blue mussels from highly lead-polluted sites at Maarmorilik into an unpolluted site and found that the mussels were only able to excrete about half of the lead originally taken up after two-three years of deployment. Zimmer et al. (2011) did a similar experiment in Ivittuut and found that blue mussels from highly lead-polluted sites had excreted only 7-21% of their original lead content after 9 months. Consequently, to evaluate metal loading at mine sites (especially at decreasing pollution), recent studies have often supplemented sampling of resident blue mussels with transplantation of blue mussels from unpolluted sites to monitoring sites, placing them in nets attached to rocks in the tidal zone and leaving them there for one year (Søndergaard et al., 2011a, Søndergaard 2013a).

In addition to this setup, 'monitoring buoys' have been tested for deployment of several transplanted organisms, including blue mussels for short time periods (weeks) (Søndergaard et al., 2014). Such 'monitoring buoys' enable measurement of bioavailable metals at a specific depth and location. Placing the mussels in nets sometimes leads to a decrease in the weight/condition of the mussels, and a direct comparison of concentrations in the mussel tissue before and after transplantation may therefore be biased. To account for this, tissue concentrations have often been converted into metal contents per mussel and the mussel nets have been placed at gradients of pollution to facilitate comparison between sites (Rigét et al., 1997). Also, since mussel metal contents vary with mussel size, the contents have often been converted into a 6 cm size mussel following the relationship reported in Rigét et al. (1997). Using this method, the difference between the metal content in the mussels before and after transplantation can be considered a proxy for the recent year's metal loading at the site. In addition to using the soft part of the blue mussels to measure bioaccumulation and metal loading, a pilot study at Maarmorilik has evaluated the possible use of the spatially-resolved chemical composition of blue mussel shells as a record of metal loading (Jessen et al., 2010). This is possible, since the shell of the mussel potentially provides a record of the chemical environment in the mussel's habitat during its lifespan (Cariou et al., 2017). Theisen (1973) reported that the lifespan of blue mussels from the Disko area on the Greenland west coast ranged from 8-12 years for a 6 cm mussel, which is a mussel size commonly found at most sites in western Greenland and south-eastern Greenland (above mid-east Greenland blue mussels appear to be absent).

Sculpins (*Myoxocephalus spp.*) have been the preferred marine fish species used in the monitoring at Greenland mine sites because it is the most sedentary of the common fish species and abundant at both western and eastern Greenland mine sites. Both shorthorn sculpin (*Myoxocephalus scorpius*) and fourhorn sculpin (*Myoxocephalus quadricornis*) are present, shorthorn sculpin being the most common (Sonne et al., 2014; Dang et al. 2017; Nørregaard et al., 2018). Sculpins are bottom-dwelling fish that can reach an age of up to c. 14 years (Søndergaard et al., 2015b) and are typically sampled by angling. As

part of the sampling, fish length, fish weight, gender and liver weight have been determined. Typically, liver and muscle have been sampled for subsequent chemical analyses to assess bioaccumulation. The liver typically contains the highest metal concentrations (Johansen et al., 1991). In addition to liver and muscle, samples of bone (Johansen et al., 1991), otoliths (Søndergaard et al., 2015b) and blood (Hansson et al., in prep.) have been used to assess the bioaccumulation of metals. A study of sculpin otoliths from Maarmorilik showed that some metals (especially lead) accumulated in the otoliths and that an otolith potentially may provide a timeline of metal exposure during the lifespan of the fish (Søndergaard et al., 2015b). However, more studies are required to explore the dynamics between metal exposure (through food and water), physiological processes (especially growth) and otolith metal accumulation in order to use sculpin otoliths as reliable records of metal exposure at Greenland mine sites.

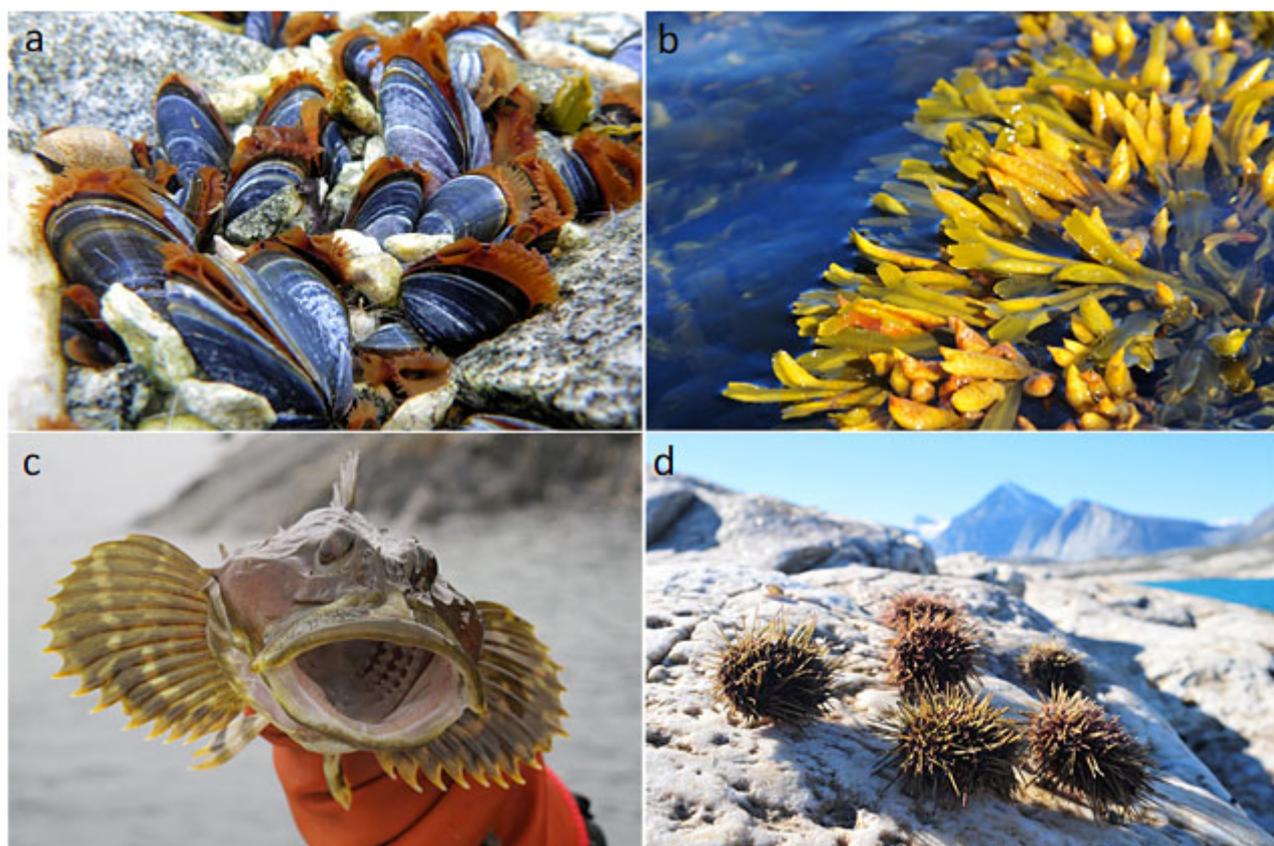


Photo 5. Bioaccumulation of pollutants in the marine environment is typically measured using: (a) blue mussels (*Mytilus edulis/trossulus*), (b) seaweed (*Fucus vesiculosus*) and (c) sculpins (*Myoxocephalus spp.*). (d) Recently, green sea urchins (*Strongylocentrotus droebachiensis*) have been tested and found adequate for monitoring purposes, especially if blue mussels are not abundant. Photo a: Ole Geertz-Hansen, all other photos: Jens Søndergaard.

3.4 Measuring dispersion and bioaccumulation of pollutants in the terrestrial and freshwater environment

Monitoring of dispersion and bioaccumulation of pollutants in the terrestrial and freshwater environment at Greenland mine sites has mainly been focused on sampling of freshwater, lichens and Arctic char (*Salvelinus alpinus*, if present) (Søndergaard and Asmund, 2011; Bach et al., 2016). In addition, sampling of surface soil, dust and freshwater sediment has been included in some previous studies (Søndergaard et al., 2012; Rambøll, 2013; Hansson et al., 2019). A recent study evaluated wolf spiders (*Pardosa glacialis* and *Pardosa*

groenlandica) as potential new monitoring organisms (Hansson et al., 2019). This is described in more detail in the following.

Sampling of freshwater in streams and lakes has been an important part of most monitoring programs and usually both filtered and unfiltered samples have been taken (sometimes with duplicates). Filtered and unfiltered samples represent dissolved and total (both dissolved and particle-bound) metals in the water, respectively. The current method for filtration of freshwater for metal analyses uses disposable 0.45 µm syringe nylon filters, a 2-component polyethylene/polypropylene syringe (without rubber O-ring) and 15 ml polypropylene vials. In addition to the water samples, *in situ* measurements of pH, temperature, electrical conductivity and redox potential (Eh)/oxygen in the water are typically performed. Sometimes, and in case of high total suspended sediment concentrations in the water, total suspended solids (TSS) have also been determined in the water by filtering a large water sample (typically 1 litre) through a filter and determining the weight of sediment collected on the filter.

Lichen (*Flavocetraria nivalis*) has been included in all monitoring programs at Greenland mine sites (except Ivituut) as indicator of dust dispersion and deposition of pollutants. The foliose lichen *Flavocetraria nivalis* has been selected because it is abundant in Greenland, has a long life span, is easy recognisable due to its yellow colour and has a great ability to concentrate pollutants deposited from the air (Rigét et al., 2000). Also, it has no roots and any metal accumulation can be attributed to atmospheric uptake. The accumulation dynamics and cellular location of lead, zinc and cadmium in *Flavocetraria nivalis* were studied by Søndergaard et al. (2013b), who showed that resident lichens at polluted sites in Maarmorilik contained significantly more lead, zinc and cadmium compared with transplanted lichens placed at the same sites after one year of deployment. This was found to be associated with a higher metal content in the 'residual' (i.e. strongly bound) metal fraction in the resident lichens (presumably due to accumulation of nearly insoluble particles near the thalli surface or in the intercellular spaces), thus demonstrating a mechanism of the lichens to cope with high metal concentrations.

Since resident *Flavocetraria nivalis* represent metals accumulated over several years, lichens transplanted from unpolluted sites into monitoring sites followed by collection the next year have been used to assess the annual dust deposition at Maarmorilik, Seqi and Nalunaq (Søndergaard et al., 2011b; Søndergaard et al., 2012; Søndergaard, 2013a; Bach and Larsen, 2016). Transplanted lichens have been placed on the ground on dead organic matter (i.e. not directly on soil) and covered by a 1×1 cm mesh nylon net held in place by small flat pieces of rocks. Typically, three c. 15×15-cm patches with lichens have been made and subsequently pooled into one sample per site. Compared with the setting up and collection of samples with conventional dust samplers, sampling of lichens requires much less time and effort and is therefore a cost-effective way to measure dust deposition from mining, especially in remote arctic areas. However, until recently, metal accumulation by lichens was only regarded as a relative (not absolute) measure of dust deposition. To address that, Søndergaard et al. (2013) compared lead, zinc and cadmium accumulation in transplanted *Flavocetraria nivalis* with absolute deposition rates using conventional bucket-type dust samplers. Based on their results, a conversion factor was derived for (rough) estimation of absolute deposition rates (at least for lead, zinc and cadmium) based on lichen transplants. Using this

method combined with lichen transplants located at numerous sites in Maarmorilik, it was possible to estimate the entire annual dust deposition of lead, zinc and cadmium in the area.

Dust deposition was measured at Maarmorilik by Søndergaard et al. (2013) using passive bucket type samplers (Bergerhoff, German Standard VDE 2119). This method involves placing open polyethylene containers in a basket on a pole 1.8 m above the ground followed by collection c. one month after. In addition, surface soil (sieved to <2 mm) has been used to assess dust dispersion and deposition at the mine sites in Maarmorilik and Seqi (Hansson et al., 2019; Søndergaard, 2019). In 2019, field investigations of real-time concentrations of atmospheric dust with subsequent elemental characterisation and source attribution have been initiated using a combined and portable optical particle counter and gravimetric dust collector (DustTrak DRX).

Freshwater bottom sediment was sampled in a lake at the ruby mine at Aappaluttoq using a Kajak sediment corer as part of the baseline study (Rambøll, 2013).



Photo 6. Dispersion and bioaccumulation of pollutants as dust in the terrestrial environment are typically assessed using (a) *Flavocetraria nivalis* lichens as they are abundant in most areas in Greenland, have no roots and take up particles/pollutants solely via the air. (b) In addition to the collection of resident lichens from the monitoring sites, lichens are sometimes transplanted to the sites, placed under nets and collected the following year to assess the dust deposition for that specific period. (c) Bergerhoff dust samples have also been used to measure dust deposition. (d) Recently, wolf spiders (*Pardosa glacialis* and *P. groenlandica*) have been successfully tested as potential biomonitors of metal pollution (Hansson et al., 2019). All photos: Jens Søndergaard.

In addition to the above, a recent study evaluated bioaccumulation in wolf spiders (*Pardosa glacialis* and *Pardosa groenlandica*) as a proxy for metal loading in the terrestrial environment at Maarmorilik (Hansson et al., 2019). The spiders were caught manually using tweezers or in yellow cups containing water with a drop of liquid detergent. Cups were dug into the ground at level with the ground surface. Collected spiders were preserved in ethanol prior to determination of species and chemical analyses.

Arctic char has been included in the monitoring program at Nalunaq due to the presence of a population of Arctic char in the Kirkespir River near the mine (Bach and Larsen, 2016). As part of the sampling, fish length, fish weight, gender and liver weight have been determined and liver samples analysed for metal concentrations to assess potential bioaccumulation of pollutants.

3.5 Measuring or assessing toxicological effects of pollutants

Most research and monitoring work conducted at Greenland mine sites have so far focused on assessing the dispersion and bioaccumulation of pollutants, while few studies have assessed the direct toxicological effects of pollutants on organisms or changes in the presence/absence of organisms related to pollutants. A description of the latter is given below.

Elberling et al. (2003) studied benthic foraminifers in sediment cores from Maarmorilik and found that metals from the mining activities (in this case mainly the submarine tailings disposal) caused significant changes in foraminifer assemblage composition at polluted sites. In addition to changes in the composition, up to 20% of the population of the foraminifer *Melonis barleeanus* at the most polluted site was deformed compared with less than 5% of the natural background population. In cores representing 100 years of sedimentation, the total number and frequency of morphological abnormalities among *M. barleeanus* revealed some correlation with heavy metal concentrations (up to $r^2=79\%$). It was therefore concluded that foraminifera abnormalities can be a useful biomarker for evaluating trends in biological impacts from heavy metal pollution.

Josefson et al. (2008) evaluated changes in benthic macrofauna composition due to submarine disposal of tailings in Maarmorilik. They found clear changes in benthic fauna composition in response to the tailings disposal, both temporally and spatially. Recolonisation 15 years after mine closure was slow and impacted areas still dominated by opportunistic species. Concentration-response relations between sediment lead concentrations and faunal indices of benthic community integrity (i.e. AMBI and DKI indices) indicated a threshold value of around 200 mg/kg, above which deterioration of faunal communities occurred. Above this threshold, diversity decreased dramatically and dominance of sensitive and indifferent species was substituted by dominance by tolerant or opportunistic species.

Sculpins are the most sedentary of the local common fish species and therefore most likely to encounter toxicological effects of pollution from point sources such as mine sites (Søndergaard et al., 2015b). Consequently, effect studies on fish have so far focused on sculpins (a few other unpublished studies have been conducted on mussels, snails and amphipods). Sonne et al. (2014) conducted histopathological studies on livers and gills of sculpins collected at five sites in Maarmorilik. Sculpins from the three most polluted sites (all located within 2 km from the mine) contained significantly more lead, mercury and arsenic in

the liver compared with the two reference sites (located 12 and 35 km from the mine, respectively). Threshold values for Lowest Observed Effect Dose (LOED) were exceeded for liver concentrations of mercury (reproduction and subclinical endpoints), arsenic and cadmium (tissue lesions, biochemistry, growth and survival). No Observed Effect Dose (NOED) levels were exceeded for liver concentrations of lead and zinc (growth, mortality and reproduction). A range of chronic lesions was observed in the sculpin livers and gills and a positive correlation was found between some liver lesions (necrosis) and gill lesions (telangiectasis) versus heavy metal concentrations. However, endoparasites occurred in highest abundances at the most polluted sites and may have been a co-factor in the development of the observed liver and gill lesions as well as indicators of the pollution.

Nørregaard et al. (2018) conducted a similar study of sculpins from two different sites in Mestersvig. Their study showed significantly higher liver concentrations of iron, mercury, manganese, lead, selenium and zinc in shorthorn sculpins at a polluted site close to the former mine compared with a reference site. Liver concentrations of mercury, arsenic, cadmium and lead exceeded reference NOED and LOED thresholds for biochemistry, tissue lesions, growth, survival and reproduction. The sculpins from the most polluted site had a significantly higher number of total gill lesions compared with the reference site. Specifically, histopathological investigations of the sculpin gills revealed significant increases in the prevalence of hyperplastic epithelium, inflammation, intensity of neutral and total mucus cells and chloride cells and increased infection of colonial Peritricha.

Dang et al. (2017) studied differences in metal liver concentrations, histopathology and presence of parasites in two different sculpin species (fourhorn sculpin (*Myoxocephalus quadricornis*) and shorthorn sculpins (*Myoxocephalus scorpius*)) also at Mestersvig. In their study, significantly higher concentrations of copper, zinc, mercury and lead were observed in the fourhorn sculpins. Also, the following histological effects: density of blood vessel fibrosis, prevalence and density of chondroplasia, number of mucin-containing mucous cells and chloride cells and mean intensity of colonial Peritricha were significantly higher in the fourhorn sculpins. This suggests that it is important to identify the species and keep the metal body burden separated per species when conducting environmental assessments.

A recent study by Dang et al. (2019) used a novel method called mucosal mapping (i.e. slime cell mapping) to study effects of metals and parasites in sculpins from Maarmorilik. The authors found that gill filament mucous cells in sculpins followed a pollution gradient and were largest and densest in fish from the most polluted site. This may have a respiratory effect on the fish due to reduction in the exposed gill surface area and increased diffusion distance between oxygenated water and the blood. The study concluded that mucosal mapping can be used as a quick, cost-efficient method to assess toxicological effects of pollutants on fish.

The studies above all focus on *in situ* toxicological effects of pollutants. Bach et al. (2014b) investigated the potential for using the arctic marine amphipod *Orchomenella pinguis* in laboratory-based water and sediment bioassays of mining pollution. *O. pinguis* was observed to effectively accumulate metals from both water and sediment, while simultaneously tolerating relatively high body concentrations of metals. Specifically, *O. pinguis* coped with high lead and zinc

body concentrations without being severely effected in terms of mortality. Consequently, if this species is to be used for bioassays in Greenland, biomarkers for effects other than mortality (e.g. feeding and burial behaviour, growth etc.) should be included. Based on the results and comparisons with literature, there was no indication that this arctic species is more sensitive to heavy metal exposure than comparable temperate or tropic species.

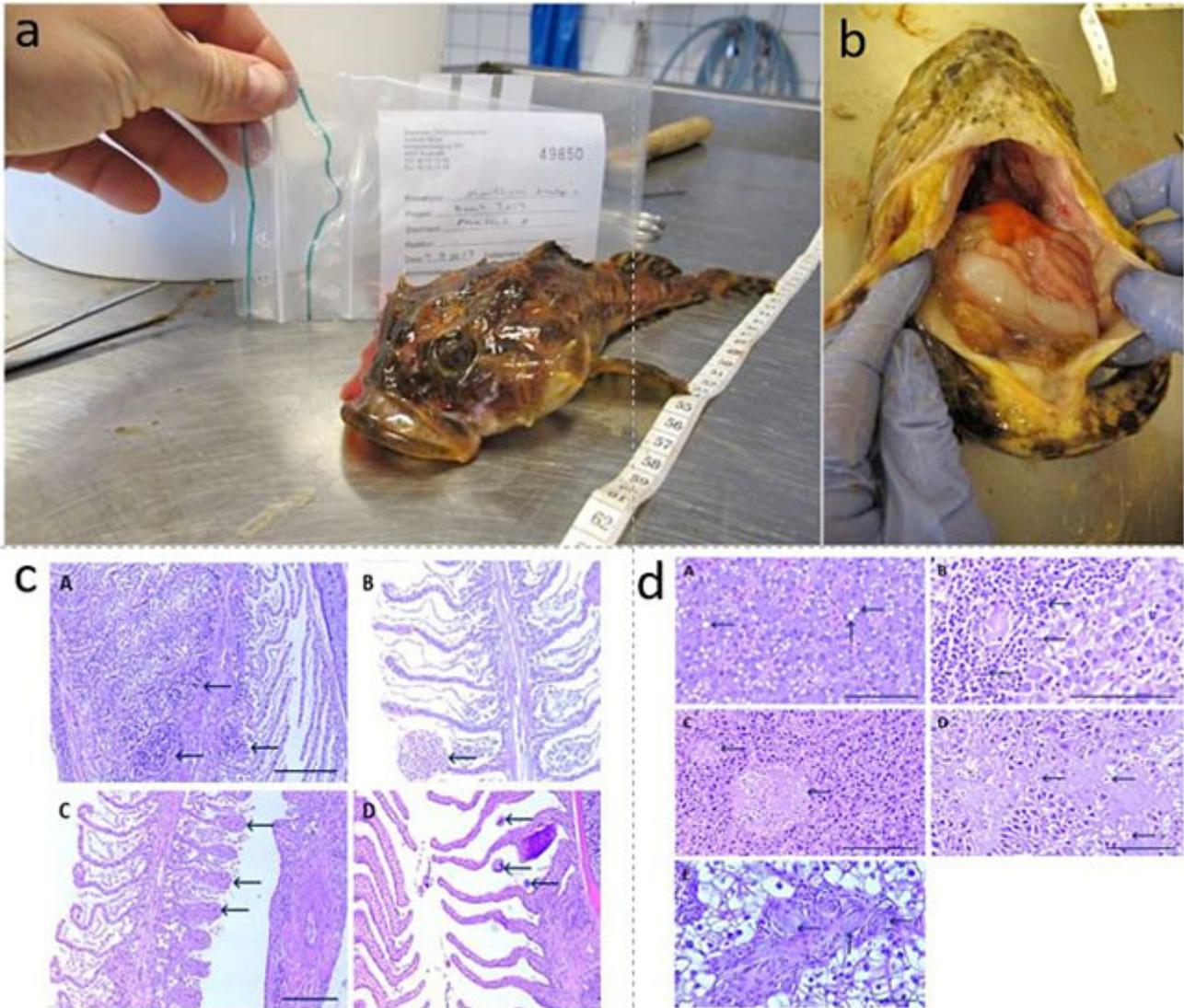


Photo 7. Most ecotoxicological effect studies at Greenland mine sites have focused on sculpins (a-b). Histopathological techniques have been used to identify lesions on (c) gill tissue and (d) liver tissue. Photo a-b: Jens Søndergaard. Photo c-d: From Sonne et al. (2014).

4 Considerations for setting up an environmental monitoring program at Greenland (and other Arctic) mine sites

Previous research and monitoring results have provided a good knowledge base for setting up an adequate monitoring program at Greenland mine sites. This knowledge can also, at least to some extent, be applied to other Arctic sites. From previous results, it is evident that the unique Arctic conditions have to be taken into account. This is especially relevant for monitoring freshwater as pronounced temporal variations in water chemistry are typically observed, for instance due to spring flushes of pollutants from weathering of minerals accumulated during winter. Also, seasonal stratigraphic mixing of seawater in the fjords can cause temporal variations in sea water chemistry, and these need to be taken into account if seawater is monitored.

Monitoring programs should always be adapted to the site- and mine-specific conditions and be designed to detect dispersion and bioaccumulation of pollutants from all potential pollution sources. The frequency and scale of the monitoring should also reflect the risk of pollution and the potential environmental impact. For instance, a mine involving exploitation of minerals containing potential environmentally problematic elements such as fluoride, heavy metals and radionuclides will require a more comprehensive monitoring program than a mine involving exploitation of more harmless rock types such as feldspar and marble. Also, the monitoring program should be dynamic and subject to continuous revision according to the monitoring results and according to changes to the mining project.

As a rule of thumb, an initial environmental monitoring program at Greenland mine sites (covering all mining phases including baseline) should at least encompass sampling and characterisation of the following: 1) freshwater from relevant streams, rivers and lakes (both unfiltered and filtered water together with measurement of pH, conductivity and total suspended solids) (see Section 3.4); 2) bottom sediment from relevant lakes and marine sites (see Section 3.2 and 3.4); 3) airborne particulates of selected particle size fractions (e.g. PM_{2.5}, PM₁₀ and/or total suspended dust) as well as total deposited dust (see Section 3.4); 4) lichens (preferably *Flavocetraria nivalis*) (see Section 3.4); 5) seaweed (preferably *Fucus vesiculosus*) (see Section 3.3); 6) mussels (preferably blue mussels, *Mytilus spp.*; typically 4-5 cm and 5-6 cm shell size intervals) (see Section 3.3); and 7) sculpins (*Myoxocephalus spp.*; liver, muscle and otoliths) (see Section 3.3). If Arctic char are present, these should be included as well (see Section 3.4). Sediment, lichens, seaweed, mussels and sculpins (and Arctic char) are usually sampled once a year, whereas more frequent water sampling may be required (especially during the operational phase of the project). For more details on sampling strategy (i.e. sampling location and frequency) and targeted pollutants, see Section 3.1. For details on sample preparation and chemical analyses, see Appendix 1.

The above applies to an initial environmental monitoring program for a small to average scale project where little to no impact is expected (although adjustments may be made, for example according to the abundance of monitoring organisms at the site). For larger projects and/or projects involving minerals of environmental concern, a more comprehensive monitoring program will be

needed, but this will not be described here. In case significant dispersion of pollutants is detected, the monitoring program should be adjusted and extended by using, for instance, more comprehensive methods as described in Section 3.2-3.5, and the scale and the frequency of the monitoring should be adjusted accordingly to evaluate the dispersion and corresponding mitigation actions.

Combination of 'chemical monitoring' (i.e. pollutants in water/sediment/air) and 'biological monitoring' (i.e. pollutants accumulated in key monitoring organisms such as lichens, seaweed, mussels and sculpins) is considered necessary since each method has its own advantages. Thus, chemical monitoring can provide total concentrations of all relevant pollutants in the environment but gives little or no information on the bioavailability and toxicity of the pollutants and the temporal variation (except for dated sediment cores). Biological monitoring can provide a time-integrated measure of bioavailable pollutants of key monitoring organisms in the habitat. Also, annual sampling of, for example, growth tips of seaweed can also provide a measure of the year-to-year variation. However, not all pollutants can be measured using biological monitoring because most organisms are, to some extent, able to regulate and excrete specific pollutants.

In addition to the environmental monitoring program described above, any significant effluent discharges (i.e. point releases of waste water, tailings etc.) associated with the project should be monitored. The frequency of the effluent monitoring should reflect the risk of pollution and the potential environmental impact. Typically, this will involve high-frequency (e.g. daily/weekly/monthly) sampling early in the project, but the frequency may be reduced later if no issues of concern are identified.

5 The role of environmental research and monitoring in minimising environmental impact of mining in Greenland

On the way to a more environmentally friendly mining industry on a global scale, adaptive monitoring of all mining activities plays a key role by linking the discharges to environmental effects. A diverse toolbox of monitoring methods is needed to be able to select efficient methods to determine discharges and specific effects of the diverse activities and environmental settings.

In Greenland, research and monitoring results have been included in the efforts to establish environmentally safe threshold limits for pollutants in seawater, freshwater and air (MRA, 2015), which have been applied in mining licenses. Furthermore, monitoring results have been used for continuous evaluation, and in some cases also regulation, of ongoing activities in order to minimise environmental impact. For example, previous environmental monitoring identified significant pollution from a large waste rock dump at Maarmorilik during the mining period, which led to a decision to remove it as part of the mine closure in 1990. However, since the waste rock dump was situated on a steep mountainside and extended down into the fjord and much of the waste rock was inaccessible or had become part of the permafrost, its complete removal was not possible. At Seqi, a dust dispersion problem was identified by the monitoring program during mine operation. In consequence, the authorities demanded initiation of actions to reduce the dust dispersion, and subsequently a dust suppressant was applied to the roads.

Also, various lessons have been learnt with regard to minimising the environmental impact at Greenland mine sites based on the results of the past research and monitoring. These include the importance of thorough geochemical leach testing of mine waste types such as waste rock and tailings prior to operation (as summed up in Søndergaard et al., 2018) and taking those results and the site-specific conditions into account when depositing the waste. Specifically, deposition of waste containing elements and minerals of environmental concern, as identified by leach testing, should not be deposited at exposed mountain slopes or in the sea (as in Maarmorilik) or in the tidal zone (as in Ivittuut). The experiences gained, together with knowledge gained from mines in other parts of the world, are now part of the knowledge base at DCE and GINR, which form the basis for the advisory work provided to the Greenland authorities on mining projects. This advisory work includes evaluation of EIAs, setting environmental requirements and conditions in licenses and more ad-hoc advising on environmental issues during the different phases of the mining operations.

Greenland has a long history of mining, starting with the cryolite mine in Ivittuut in 1854 and with significant mining operations at Mestersvig in the 1950-60s and Maarmorilik in the 1970-80s (Table 1). At these three mine sites, no or only extremely few environmental investigations were conducted prior to the operation – no EIAs were made and little attention was paid to the potentially adverse environmental consequences of mining activities and waste deposition methods. Thus, those mine sites have left a legacy of long-lasting pollution (Table 1). In contrast, no significant chemical pollution can be traced in the areas of Nalunaq, Seqi, Aappaluttoq and White Mountain where mining

was/is undertaken in the 2000-2010s. This historical trend is considered, at least in part, to be due to the development of a regulatory system with requirements for thorough EIAs and implementation of strict environmental requirements and conditions in licences, based on the highest international standards and the knowledge obtained from legacy mines in Greenland and elsewhere.

6 Future perspectives

In the coming years, there are plans to supplement monitoring at mine sites at local scale in Greenland with more extensive regional monitoring in selected areas, including also assessment of cumulative effects (e.g. disturbance from increased shipping, helicopter transport etc.) in the areas of concern. Furthermore, there is a potential to involve the locals near a mine site in the monitoring to take advantage of local ecological knowledge and to build local confidence to the environmental protection and ensure a social “license to operate” for mining companies. This is beneficial for establishing a mining industry (involving also ‘small-scale mining’) in areas where the local population is closely connected to the landscape and where both the economy and food security are based on local fishing and hunting. With regard to future research, knowledge gained from past and current research at mine sites in Greenland and elsewhere, along with the ongoing development of mine sites in Greenland and the environmental challenges they pose, will form the basis for the direction of the research. In addition, technological advances and applied research constantly create new opportunities to improve and extend methods for environmental monitoring at mine sites. The following list covers examples of research topics related to environmental monitoring that could be relevant for Greenland mine sites in the coming years.

- High-spatial-resolution soil surface and sediment mapping of trace metal contents using field-portable handheld XRF.
- Dust monitoring using field-portable instruments (e.g. DustTrak) for measuring total air dust concentrations in various size fractions as well as the chemical composition of dust particles using filter collection.
- Evaluation of methods to mitigate dust dispersion at mine sites.
- Spatial trace metal analyses of solid biogenic materials (otoliths, bones, shells, plant tissues) as archives of pollution (using laser ablation ICP-MS analyses).
- Fish toxicology as a response to mining pollutants.
- Genetic studies of environmental samples (commonly referred to as eDNA studies) to evaluate changes in biodiversity near Greenland mine sites.
- Evaluation of more species for use in the monitoring of mining pollution (covering more locations, sources for uptake and trophic levels).
- Radioactive elements in the Greenland environment, especially near Kvanefeld in South Greenland.
- Identification of mining pollutants using stable isotopic signatures.
- Speciation of pollutants and characterisation of mine waste in terms of potential toxicity and methods to reduce leaching/toxicity.

7 Final remarks

Environmental monitoring plays a key role in minimising the environmental impact of mining. In Greenland, some of the old mines have a legacy of long-lasting pollution and practices for environmental monitoring have evolved over the years following extensive research conducted at these former mine sites. Monitoring results at Greenland mine sites have formed the basis for decision-making and regulation of mining activities and have assisted in reducing adverse environmental effects in several cases. From the Greenland monitoring and research results, it is evident that monitoring programs should always be adapted specifically to the mine site of concern. Adapted monitoring programs should include a diverse suite of monitoring organisms among abiotic techniques and take into account temporal variations in discharge of pollutants unique to the Arctic. The future environmental monitoring of mining activities in Greenland is likely to include a range of additional tools for assessing environmental impact owing to the continued research in mining areas and the development of new techniques. Future monitoring programs may also include results from regional monitoring programs and assessment of cumulative effects of mining activities on a larger scale.

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9 Appendices

Appendix 1. Methods for sample preparation and chemical analyses

Most analyses of environmental samples from mines in Greenland have been performed at the accredited Trace Metal Laboratory at Danish Centre for Environment and Energy (DCE) in Roskilde, Denmark. The laboratory is accredited by the Danish Accreditation Fund (DANAK) following ISO 17025 to analyses of biota, sediment, fresh- and seawater for a range of elements (Accreditation no. 411). The following describes the current methods and practices for sample preparation and chemical analyses of non-radioactive elements.

With regard to storage, in general biota, sediment and soil samples are collected and stored in polyethylene bags and frozen at -20°C until analyses. An exception is samples for PAH (i.e. oil components) analyses, which are sampled in RILSAN bags and frozen.

Water samples are usually taken in polyethylene bottles or vials and stored cold at 5°C or at room temperature. After reception of the water samples in the lab, a clean acid (i.e. Suprapure- or Ultrapure-grade nitric acid; 1 ml/l for fresh water and 2 ml/l for seawater) is added to preserve the samples (i.e. to keep metals from precipitating and dissolve metals that may have precipitated). Also, a series of at least five 'blank samples', associated with the specific batch of samples are made, using Milli-Q water and the same acid as for the samples.

Sediment, soil and most biota samples are freeze-dried prior to the chemical analyses. Oven-drying is avoided to minimise the risk of mercury loss from the samples. After freeze-drying, biota samples (seaweed, mussels etc.) are typically homogenised to powder in an agate mortar to enable to take out a representative subsample. Exceptions are lichen samples, which are kept as they are without homogenisation, and small samples (like spiders) composed of less than 300 mg dry weight, which is the sample weight typically used for analyses. Samples of liver and muscle are most often not freeze-dried but instead cut directly out of the main sample after thawing (avoiding the originally exposed surfaces on the main sample).

For most analyses of sediment, soil and biota, sample digestion is needed. The most frequently used digestion method involves a subsample (300 mg dry weight or 1 g wet weight biota and 200 mg dry weight sediment/soil) digested in half-concentrated Suprapure nitric acid in a microwave oven (according to Danish Standard, DS 259). For sediment and soil, sometimes a more rough digestion method is applied using a mixture of hydrofluoric, hydrochloric and nitric acid in Berghof bombs at 120°C in a heating oven for 4 h followed by neutralisation of the hydrofluoric acid with boric acid (Søndergaard et al., 2011a). For each batch of digestions (which for the current system involves 18 samples), one 'blank sample' (i.e. a vial exposed to the same treatment as the samples but without sample content), one duplicate sample and 1-3 different Certified Reference Materials (CRMs), preferable of the same type as the samples, are included in addition to the samples. After digestion, the solutions are further diluted with Milli-Q water prior to analyses.

Most analyses are performed using inductively coupled plasma mass spectrometry (ICP-MS) (currently an Agilent 7900), which enables simultaneous analyses of more than 60 elements, including those of typical environmental concern with respect to mining. Freshwater and digestion solutions of sediment, soil and biota are analysed directly on the ICP-MS. The currently established main method for freshwater, sediment, soil and biota determines the following elements: Li, Be, Na, Mg, Al, P, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Rb, Sr, Y, Zr, Nb, Mo, Ru, Pd, Ag, Cd, Sb, Te, Cs, Ba, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Hf, Ta, W, Re, Pt, Au, Hg, Tl, Pb, Bi, Th and U (using the elements Ge, In, Rh and Ir as internal standards). Seawater has previously been analysed for a selection of elements (V, Mn, Co, Ni, Cu, Zn, Cd and Pb) using pre-concentrations of the metals on a column filled with a chelating agent (Chelex-100) followed by elution of the metals using nitric acid (Søndergaard et al., 2015a). However, a new cost-efficient method for seawater analyses using Agilent 7900s Ultra High Matrix Introduction system coupled to Agilent's ISIS-3 sample introduction system has recently been developed for analyses of selected elements (V, Mn, Ni, Cu, Zn, Ag, Cd, Pb and U).

Additional analyses include mercury analyses using a dedicated instrument for direct analyses without prior sample digestion (a Milestone DMA-80).

With respect to data analyses, element concentrations in samples are determined by subtracting 'blank' values from the sample results, and duplicates and CRMs are used to estimate the precision and accuracy of the results. Variations on the 'blank' samples are used to determine the analytical Detection Limit (DL) (usually as 3 standard deviations on a series of blank samples). Finally, sample concentrations are compared with the DL and reported as being below the DL, when appropriate.

With regard to analyses of radioactive elements, DCE has recently established a radioecology laboratory facility with the necessary approval from the Danish Health Institute, and the current instrumentation includes an Ortec Alpha Spectrometer (used for measurements of ^{210}Po , ^{210}Pb , ^{228}Th , ^{230}Th , ^{232}Th , ^{234}U , ^{235}U and ^{238}U) and a Colibri very low dose VLD-100 SAB meter (for measurement of gamma dose rates, alpha and beta contamination check) from Canberra/Mirion. The lab is under further development and a gamma spectrometer will be installed in the near future.

As part of the QA/QC procedure, the Trace Metal Laboratory at DCE is subjected to regular internal and external audits and participates in the international laboratory intercalibration program for marine environmental samples QUASIMEME (www.quasimeme.org) twice a year. This involves reporting of chemical results of unknown samples of marine biota, sediment and seawater followed by an evaluation based on the reporting of corresponding results from a range of other laboratories in Europe.

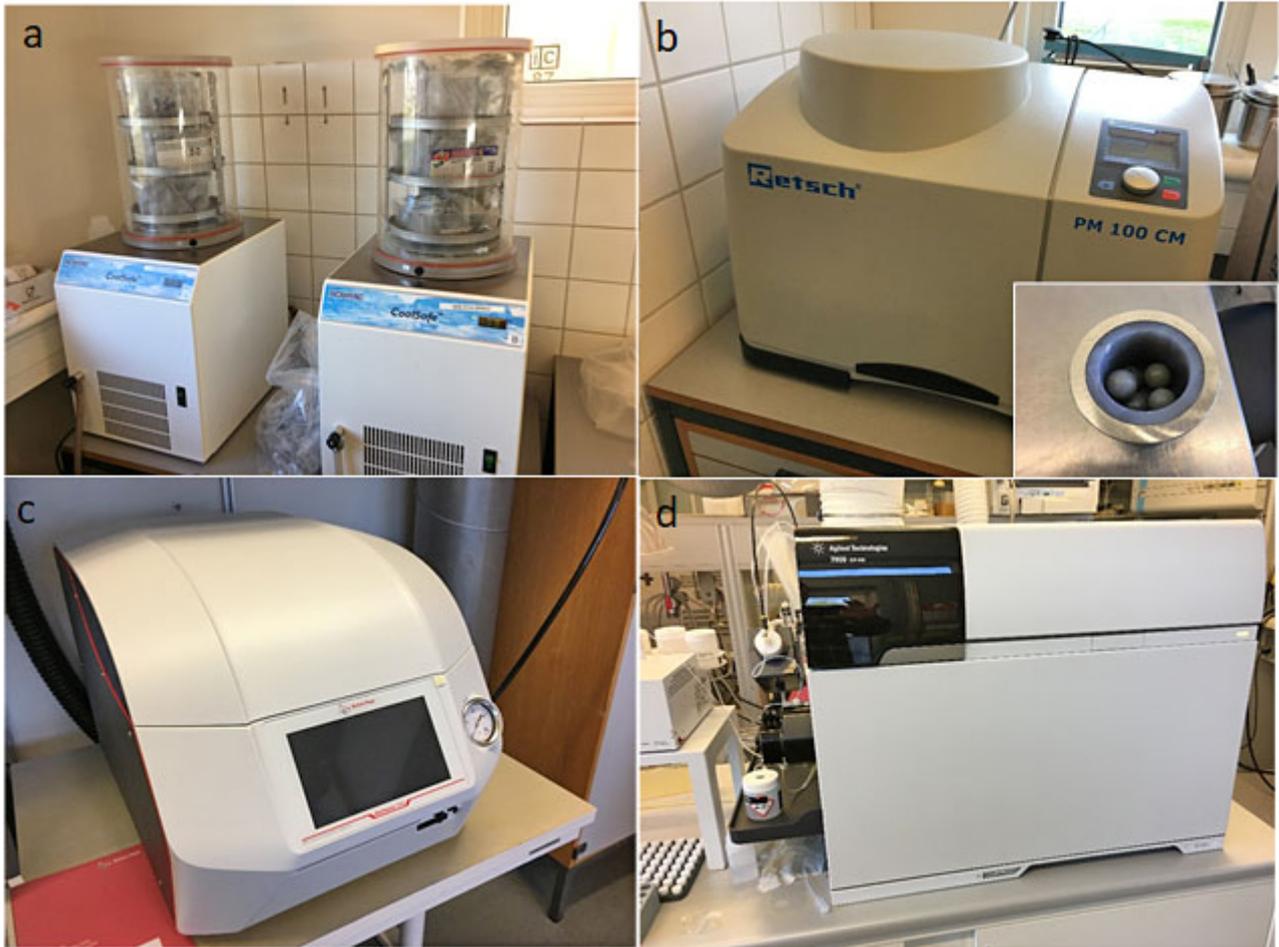


Photo 8. When samples are received at the accredited Trace Metal Laboratory at DCE, most are: (a) freeze dried, (b) homogenised in an agate mortar (for mussel tissue and seaweed), (c) digested in acid in a microwave oven and d) measured using an Inductively Coupled Plasma Mass Spectrometer (ICP-MS) for 60+ elements. All photos: Jens Søndergaard.

Appendix 2. Practices for storage of samples and data

At DCE, a large -20 °C freezer serves as a sample bank that DCE administers for the Greenland authorities. The sample bank contains samples collected from Greenland mine sites in the past before mining operations began, so-called baseline samples that can be analysed for control at any time. This could be relevant in case of a pollution event and especially for detection of pollutants not previously analysed for in the samples. The sample bank also includes samples from the monitoring programs undertaken at current and former mine sites as well as research projects associated with these.

All data derived from samples gathered in connection with the environmental monitoring at Greenland mine sites are entered into a database that DCE administers for the Greenland authorities. The data are available and used in the advisory work that DCE, in collaboration with the GINR, provides to the Greenland authorities.

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ENVIRONMENTAL MONITORING AT MINE SITES IN GREENLAND

A review of research and monitoring practices and their role in minimising environmental impact

This report provides an overview and status of environmental research and monitoring practices near mine sites in Greenland. The most significant mines in Greenland's mining history are described together with research results and monitoring practices used for assessing dispersion, bioaccumulation and toxicological effects of mining pollutants. This knowledge is used to provide recommendations for setting up an adequate environmental monitoring program. Finally, the role of environmental monitoring in regulating mining activities and minimising the associated environmental impact is discussed along with future perspectives and directions for research and monitoring near Greenland mine sites. The report is considered especially relevant for researchers, advisors, mining companies/consultants and people working with the administration of environment and mining in Greenland. However, many of the conclusions and recommendations can also be applied in environmental monitoring and regulation of mining and other activities in other countries, and the report is therefore relevant to a broader audience.