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Review

# Mining pollution in Greenland - the lesson learned: A review of 50 years of environmental studies and monitoring

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## HIGHLIGHTS

- Three legacy mines in Greenland caused metal pollution and areas are still polluted.
- No significant pollution was identified at mines established in the last two decades.
- A comprehensive EIA process and adaptive monitoring are essential.
- Monitoring must be site-specific, diverse and take Arctic conditions into account.
- Climate change underscores the importance of ongoing monitoring and mitigation.

## GRAPHICAL ABSTRACT

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## ABSTRACT

This review provides an overview of environmental studies and monitoring at mine sites in Greenland since the first environmental studies were conducted in the early 1970s. Mining at three legacy mine sites in Greenland (Ivittuut, Mestersvig and Maarmorilik) caused significant metal pollution, mostly with lead and zinc, due to lack of adequate environmental studies and regulation. These legacy mine sites have later served as study areas for development of methods for environmental monitoring, which can also be applied to other sites. The review describes the most significant mines in Greenland's mining history together with procedures for conducting the environmental monitoring work. A comprehensive description is provided on the research results and development of monitoring practices during the past 50 years for assessing dispersion, bioaccumulation and toxicological effects of pollutants in both the marine and terrestrial environment. Further, the current practices for sample preparation, chemical analyses and storage of samples and data are described. From the studies it is clear that monitoring needs to be site- and mine-specific, adaptive, diverse and take conditions unique to the Arctic into account, such as permafrost, seasonal drainage and fjord stratification dynamics. Based on the results, lessons learned for future monitoring programs are given. Moreover, spatial and temporal trends of the legacy pollution at the Greenland mine sites are discussed. Finally, it is shown how research and monitoring results have been applied to regulate mining activities in Greenland to minimise the environmental impact, and some future perspectives are presented. Many of the results and conclusions in the review are considered applicable to environmental monitoring of mining and other industrial activities in other areas than Greenland, both inside and outside of the Arctic.

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**Contents**



## 1. Introduction

Greenland has a long mining history starting with industrial extraction of cryolite in 1854 in Ivittuut in South Greenland (Johansen et al., 2010b). However, it was not until the early 1970s that the first environmental studies were conducted prior to opening of the lead-zinc mine in Maarmorilik in West Greenland (Asmund et al., 1994). Like many other countries, Greenland has a legacy of long-lasting pollution from former 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, resulted in significant environmental pollution, mostly with lead and zinc (Johansen et al., 2010b; Søndergaard et al., 2011a; Aastrup et al., 2018). These polluted legacy mine sites have enabled studies of dispersion, bioaccumulation and toxicological effects of mining pollution under Arctic conditions during the past 50 years and have provided valuable information for the development of a regulatory system with Environmental Impact Assessments (EIA) and monitoring guidelines to minimise the impact of new mining activities. Since the 1970s, monitoring of Greenland mine sites has been performed regularly at both operating and closed mines. In addition to the monitoring activities, numerous environmental studies have been carried out at the legacy mine sites over the years by different research groups.

This review aims to provide a thorough overview of environmental research and monitoring studies undertaken at mine sites in Greenland and sum up conclusions from the results gained during the 50 years of research and monitoring. First, the most significant mines in Greenland's mining history are described along with their environmental characteristics. Procedures for conducting monitoring and advisory work, specifically the roles of the mining companies and their consultants versus those of the authorities and their advisors, are described. A comprehensive description of previous research and monitoring studies is presented for assessing dispersion, bioaccumulation and toxicological effects of pollutants from the Greenland mine sites, and the most important findings are highlighted. In addition, current methods for sample preparation, chemical analyses and storage of samples and data are described. Based on this knowledge, lessons learned for future monitoring programs are presented. Spatial and temporal trends of legacy pollution at Greenland mine sites are subsequently summed up and discussed. Finally, it is highlighted how environmental research and monitoring results have been applied to minimise environmental impacts in Greenland, and future perspectives for environmental monitoring at Greenland mine sites are given.

## 2. Former and current mine sites

The location of former and current mine sites in Greenland is shown in Fig. 1, and more detailed information on the mines is provided in Table 1. Greenland includes a large ice-free land area of about 410,000 km$^2$, spanning over more than 20 degrees latitude, while the Greenland Ice Sheet covers around 80% of the total area. Greenland is very thinly populated with about 57,000 inhabitants (in 2021), and there are no roads to connect the cities, towns and settlements. It is a general feature of mines in Greenland that they are located close to the sea or fjords.

### 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 that is used as a flux agent in the electrolytic process of aluminium extraction from the aluminium-rich oxide ore bauxite, and the mine at Ivittuut was the only major cryolite mine in the world. 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 t 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, and the rocks were thus saturated 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 dissolution and transport of lead and zinc from the waste rock (present in the sulphide minerals galena and sphalerite) situated at the coastline. In 1985, it was estimated that between 400 and 1000 kg dissolved lead from the waste rock entered the Arsuk Fjord. 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 did not have elevated concentrations of heavy metals. Consequently, the annual environmental studies carried out later at Ivittuut in the

1980s and every two-three years during the period from 1990 to 2013 had focus on these two species (Bach et al., 2014a).

### 2.2. The lead-zinc mine at Mestersvig, East Greenland

The lead-zinc mine at Mestersvig in East Greenland was in operation only 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 t ore were produced, yielding 58,000 t lead concentrate and 75,000 t zinc concentrate. Lead and zinc were contained as sulphide minerals (galena and sphalerite) 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 subsequently loaded and shipped.

The first environmental study at Mestersvig was conducted in 1979, and the results revealed significant pollution with especially 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, which were 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., 2018). Seals in Kong Oscars Fjord did not show elevated heavy metal levels compared with elsewhere in Greenland (Johansen et al., 2008).

### 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, and the mine entrances were located at 600 m altitude on the Black Angel Mountain and connected to a mining town at sea level by a 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 mainly marble and pyrite. The ore was transported to the mining town, where it was processed into concentrate, loaded and shipped. The total production during 1973–90 was 11.2 million tonnes ore, resulting in 590,000 t lead concentrate (70% lead, 420 ppm silver) and 2,327,000 t zinc concentrate (58% zinc) (Thomassen, 2003).

The mountainous topography prevented the establishment of a land-based tailings disposal system. The tailings from the ore processing were therefore discharged into the small (2 $km^2$) fjord Affarlikassaa, which is partly separated from the outer fjord Qaamarujuk 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 t 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 Affarlikassaa Fjord (Asmund, 1992a).

The first environmental studies at Maarmorilik were initiated in the early 1970s and 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, mercury, copper and arsenic, 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, 2011b; Sonne et al., 2014; Hansson et al., 2019; Hansson et al., 2020). During the mining period, the tailings and the waste rock as well as dispersion of metal-laden dust acted as significant sources of pollution. Disposal of tailings into the Affarlikassaa 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 Affarlikassaa Fjord to the Qaamarujuk Fjord as a result of complex hydrographical processes (Asmund, 1992b; Poling and Ellis, 1995). The key hydrological processes for dispersion of pollutants from the Affarlikassaa Fjord were seasonal vertical mixing of the water layers in the Affarlikassaa Fjord and complete flushing of pollutants from the 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 Affarlikassaa 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.

### 2.4. Other mine sites

The cryolite mine at Ivittuut and the lead-zinc mines at Mestersvig and Maarmorilik described above are the most important of the former mines in Greenland from an environmental perspective. At these sites, significant pollution of the environment occurred, resulting in frequent environmental monitoring and numerous studies and assessments. There have been other mines in the Greenland mining history, as briefly described below, but without any significant pollution to the surrounding environment. That said, the old (pre-1970s) mine sites were not monitored during operation, and there may have been some pollution during that time.

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, which operated from 1924 to 1972 and produced 570,000 t

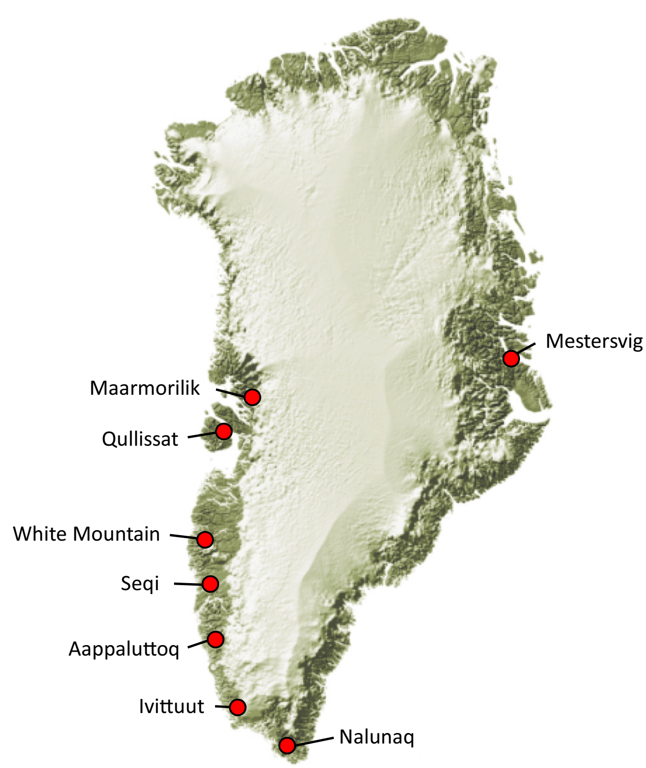


**Fig. 1.** Map of Greenland with major former and present mine sites.

**Table 1**
Major former and present mine sites in Greenland with some environmental characteristics (at the time of writing in 2021) and with references to specific studies conducted at these sites listed in the review.

| Mine | Ore type | Mining period | EIA | First environmental monitoring/study | Major environmental issue(s) | Environmental legacy | Studies referenced |
|---|---|---|---|---|---|---|---|
| Ivittuut | Cryolite | 1854–1987 | No | 1982 | Marine pollution in the Arsuk Fjord with mainly lead and zinc caused predominantly by leaching from waste rock situated in the tidal zone along the coastline and between the mine pit and the Arsuk Fjord. In addition, some input of lead and zinc from mine pit water to the Arsuk Fjord. Elevated concentrations were observed in mussels and seaweed but not in other monitoring organisms like fish and prawns. The maximum spatial extent of the impacted area during the mining period is unknown but encompasses an area at least 10-15 km from the mine. | The Arsuk Fjord system was still polluted with lead and zinc during the last study in 2013 with elevated concentrations observed within a distance of c. 5 km from the mine. A decreasing pollution trend has been observed since the first environmental study in 1982. During the period 1982–2013, concentrations decreased by roughly a factor of 3 for lead and roughly a factor of 2 for zinc measured in mussels and seaweed. | Bach et al. (2014a); Johansen et al. (2010b); Zimmer et al. (2011) |
| Qullissat | Coal | 1924–1972 | No | 2015 | None identified[a]. | None identified[a]. | Søndergaard et al. (2017) |
| Mestersvig | Lead-zinc | 1956–1963 | No | 1979 | Marine and terrestrial pollution in Kong Oscars Fjord and the Mestersvig area with mainly lead and zinc (and some cadmium and copper) from: 1) a tailings deposit near Tunnel River immediately downslope from the mine situated c. 10 km inland from the harbour; 2) dust dispersion of concentrate along the haul road; 3) spills of concentrate at Nyhavn (i.e. the harbour) and later a collapse of the quay between 1986 and 1991. Elevated concentrations were observed in sea- and freshwater, soil, sediments, lichens, vascular plants, seaweed, bivalves and sculpins (not in seals). The maximum spatial extent of the impacted area during the mining period is unknown but covers an area at least 10–15 km from the pollution sources. | The Mestersvig area was still polluted with mainly lead and zinc during the last study in 2014. Elevated concentrations were observed near the mine, along the haul road, at Nyhavn and in the adjacent fjord at a distance of at least 5 km from pollution sources. A general trend toward decreasing concentrations of pollutants has been observed since 1979 but with a peak in 1991. During the period 1979–2014, concentrations of pollutants decreased by roughly a factor of 3 for lead and zinc at Nyhavn as indicated by measurements in seaweed. | Aastrup et al. (2018); Asmund et al. (1996); Dang et al. (2017); Nørregaard et al. (2018) |
| Maarmorilik | Lead-zinc-silver | 1973–1990 | No | 1972 | Marine pollution in the Affarlikassaa Fjord and the Qaamarujuk Fjord system with mainly lead and zinc (and some cadmium, mercury and other elements) from: 1) tailings deposited in the Affarlikassaa Fjord (a small sill fjord) in immediate vicinity to the mine; 2) waste rock deposited on mountain sides downslope from mine entrances and in the fjords; 3) dispersion of metal-laden dust associated with ore extraction, transport, processing, loading of concentrate etc. Elevated concentrations were observed in seawater, sediments and a long range of marine organisms as well as in soil, lichens and spiders in the terrestrial environment. The maximum spatial extent of the affected area during the mining period is unknown, but dust from the mine was measured in lichens at least 40 km from the mine. Elevated concentrations of pollutants were measured in the Qaamarujuk Fjord system in seaweed and blue mussels at least 30 km from the mine. | The Affarlikassaa and Qaamarujuk fjords were 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 deposition of mainly lead, zinc and cadmium was still significant at a distance of at least 12 km from the mine, presumably derived mainly from continuously degrading waste rock at the mountain slopes and from residues from the mining town. The pollution decreased markedly after the mine closure in 1990. Lead and zinc levels in blue mussels and seaweed indicated that the pollution near the mine had decreased by roughly a factor of 10 for lead and a factor of 2–3 for zinc after it peaked during the mining period. Since around 2000, the concentrations of pollutants have remained relatively stable in the area with no clear decreasing trend. | Asmund (1992a, 1992b); Asmund et al. (1991, 1994); Bondam (1978); Dang et al. (2019); Elberling et al. (2002, 2003); Hansson et al. (2019, 2020); Jessen et al. (2010); Johansen et al. (1991, 1997, 2006, 2008, 2010a); Josefson et al. (2008); Larsen et al. (2001); Lewis and Perkin (1982); Loring and Asmund (1989); Møller (1978); Møller et al. (1982); Møller and Pedersen (1983); Perner et al. (2010); Poling and Ellis (1995); Rigét et al. (1997); Schiedek et al. (2009); Sonne et al. (2014); Søndergaard et al. (2010, 2011a, 2011b, 2013, 2014, 2015b, 2019); Søndergaard (2013b); Thomassen (2003); Wenne et al. (2016) |

**Table 1** (*continued*)

| Mine | Ore type | Mining period | EIA | First environmental monitoring/study | Major environmental issue(s) | Environmental legacy | Studies referenced |
|---|---|---|---|---|---|---|---|
| Nalunaq | Gold | 2004–2013 | Yes | 1998 | None identified. Cyanide was used in the production, but no significant dispersion of cyanide was observed in the environmental monitoring. | None identified. | Bach (2020) |
| Seqi | Olivine | 2005–2009 | Yes | 2004 | Dispersion and deposition of mining-related dust were measured in the terrestrial environment at Seqi during the mining period. This was indicated by elevated concentrations of chromium, nickel, iron and cobalt in lichens measurable at distances up to 5 km from the mine. The dust deposition decreased after 2008 following actions to reduce the dust dispersion from the roads using a dust suppressant and later closure of the mining activities in 2010. | None identified. During 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 very locally (within c. 1 km of the mine). | Søndergaard and Asmund (2011); Søndergaard (2013a, 2019) |
| Aappaluttoq | Rubies | 2017-date | Yes | 2006 | None identified. | None identified. | Rambøll (2013) |
| White Mountain | Anorthosite | 2018-date | Yes | 2012 | None identified. | None identified. | Søndergaard and Jørgensen (2021) |

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

coal. A recent study conducted in 2015 evaluated the environment at Qullissat and found no indication of significant chemical pollution (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 2005 to 2009; 3) a ruby mine at Aappaluttoq in Southwest Greenland, which opened in 2017; 4) a feldspar (anorthosite) mine at 'White Mountain' in Southwest Greenland, which opened in 2018. The ruby mine and the anorthosite mine are in operation at the time of writing (2021). No significant environmental impact has been identified at these mines (Søndergaard, 2019; Bach, 2020), which is considered mainly due to a comprehensive EIA process followed by environmental regulation and frequent and comprehensive environmental monitoring and control.

## 3. Environmental monitoring and advisory tasks

Since the 1970s, the working group at Danish Centre for Environment and Energy (DCE) at Aarhus University has conducted environmental monitoring at Greenland mine sites for the Greenland authorities (under changing institution names, previously National Environmental Research Institute, Greenland Environmental Research Institute and Greenland Fisheries and Environmental Research Institute). Reports from the monitoring are available to the public. In the last decade, the environmental monitoring has been done in collaboration with the Greenland Institute for Natural Resources (GINR). Historically, monitoring at active mine sites in Greenland has consisted of self-monitoring by the mining companies combined with audit monitoring by DCE and GINR on behalf of the Greenland authorities. The self-monitoring has included high-frequency water sampling and tailings sampling at outlets from the mining site. Audit monitoring has typically been based on a wide range of samples collected on an annual basis in the mining area and in adjacent areas. Monitoring after mine closure has previously been conducted entirely by DCE and GINR on behalf of the authorities. Lately, consultant companies hired by the mining companies have taken over part of the monitoring work previously done by DCE and GINR.

The methods and techniques for environmental monitoring have evolved over the years concurrently with technological advances and the 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 obtain more knowledge of the impacts. DCE and GINR are advisors to the Greenland authorities on all environmental issues related to mining from prospecting to after mine closure. Further, DCE and GINR hold and administer a large sample- and databank for the Greenland authorities with environmental samples and data from former and present mine sites, which is used in the regulation of mining activities and for research purposes. DCE has an ISO 17025 accredited trace metal laboratory and a radioecology laboratory used for chemical and radiological analyses of environmental samples whose results provide the basis for advisory and assessment.

Before a mining company can be granted a license for exploitation in Greenland, a survey of the environmental conditions at the site prior to the activities is required, and this will later form the basis for the monitoring of environmental changes. 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. These baseline results form part of the obligatory Environmental Impact Assessment (EIA) report, which assesses the expected environmental impacts of the project. According to the current practice, two-three years of baseline data are needed (MRA, 2015). DCE and GINR have advised on the development of specific guidelines for the EIA work including environmental programs and a set of water and air quality criteria for Greenland mining activities (MRA, 2015). DCE and GINR are subsequently involved in the assessment of the EIA for the authorities. A correct and fulfilling EIA is one of the reports needed before the Greenland Self-Government can decide for or against a specific mining project.

## 4. Experience from past research and monitoring studies

### 4.1. Overall sampling strategy and targeted pollutants

Past environmental monitoring studies at Greenland mine sites have been based on the collection and analyses of samples from a range of sampling stations established in the fjords, along the coastline and inland. Typically, a total of 25–50 sampling stations have been used. Most sampling stations have been placed near the potential sources of pollution to most accurately assess pollution levels and identify sources, while the remaining sampling sites have been located in a gradient away from the mine. The dominant directions for dispersion by wind or currents have been taken into account by placing more stations in the dominant directions. If freshwater samples have been taken in streams or lakes, the sampling stations have typically been placed both upstream and downstream from the potential pollution source and at the dominant lake inlets and outlets. Seasonal variations in water chemistry can be very pronounced in Greenland and other places in the Arctic, for instance due to spring flushes of winter-accumulated pollutants on land released during thaw and snowmelt (Elberling et al., 2007; Søndergaard et al., 2007; Søndergaard et al., 2012). The sampling periods for freshwater are therefore set to cover the expected variability. In most studies, reference samples from one or two sampling stations located far away from the mine (and therefore unlikely to be influenced by the mining activities) have been used for comparison. At recent mines, where environmental baseline studies have been made prior to the actual mining, sampling stations have typically been re-used in further monitoring studies because this allows the most accurate comparison of sample composition before and after mining. Finally, accessibility is an important parameter, and the sampling stations have most often been placed to permit access by foot or boat to avoid the high cost of helicopter transportation.

Regarding the targeted pollutants, focus has typically been 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 an adequate 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 are: Maarmorilik: lead, zinc, cadmium and mercury; Ivittuut: lead and zinc; Mestersvig: lead, zinc, cadmium and copper; Seqi: nickel, chromium, iron and cobalt. At the former gold mine in Nalunaq, cyanide used for ore extraction was monitored frequently besides elements such as arsenic, copper, chromium and cobalt, which are present in the ore. 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).

### *4.2. Assessment of dispersion of pollutants in the marine environment*

Since the first environmental studies at the Black Angel mine at Maarmorilik 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 (c. every 5 or 10 m) from the surface to the bottom using metal-free water samplers (like 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 in the 1970s and 1980s at the Black Angel mine where 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) measuring 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; Hansson et al., 2020). 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) but require a setup to keep them mounted and suspended in the water column during the deployment period, for instance using a buoy setup with an anchor to the seafloor (Søndergaard et al., 2014). DGT samplers only work for some metals depending on the type of the sampler (www.dgtresearch.com). 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 have been used to evaluate the dispersion, sedimentation rate and impact of pollutants on the seafloor using either grab or core samples (Loring and Asmund, 1989; Elberling et al., 2002; Perner et al., 2010; 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 c. 1 m, but they are heavy and require a larger ship and a winch. At Maarmorilik, Loring and Asmund (1989) used layers of a core sample applying $^{210}$Pb (lead-210) radiometric dating to assess the sedimentation rate of polluted sediment. Elberling et al. (2002) utilised the same approach at Maarmorilik but took it 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 during a 100-year period. Furthermore, Elberling et al. (2002) employed ratios between pollutants (here 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 source trace and separate lead from the mine from the natural background lead in the area. Stable isotopic signatures of lead were later used to identify lead from the mine versus the natural background in lichens, seaweed and blue mussels that were used as key monitoring organisms at Maarmorilik (Søndergaard et al., 2010). For sediment, a difference in grain size between sites or within a core can bias a direct comparison (Quevauviller et al., 2011). To normalise for differences in grain size of the sediment within/or between cores or grab samples, the ratio between the pollutant and aluminium (regarded as a conservative tracer) has been applied in a number of studies to improve the basis for comparison (Perner et al., 2010: Søndergaard et al., 2011a). In other studies, sediment has been sieved to a certain fraction (e.g. <2 mm or <0.063 mm) prior to analyses to improve the comparison (Asmund et al., 1991; Søndergaard et al., 2019; Hansson et al., 2020).

In addition to sampling and measurements of water and sediment, oceanographic studies have been performed to obtain knowledge of currents, mixing and dispersion of water masses and pollutants in the marine environment. In such studies near the Black Angel mine at Maarmorilik, 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 m depth in the small Affarlikassaa Fjord that is separated from the larger Qaamarujuk Fjord system by a sill at c. 20 m depth. During summer, the saline-polluted bottom water in Affarlikassaa Fjord was stagnant and separated 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 highly 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 the Affarlikassaa Fjord from the Qaamarujuk Fjord as a bottom current replacing the existing bottom water, which led to complete flushing of suspended or dissolved pollutants from Affarlikassaa Fjord to the outer Qaamarujuk Fjord system.

*4.3. Assessment of 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 assessing bioaccumulation in the marine environment near mine sites in Greenland (Johansen et al., 1991; Søndergaard et al., 2011a). Further, bioaccumulation in a range of other species has been studied, including 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*) (Johansen et al., 1991; Larsen et al., 2001; Søndergaard et al., 2014). A recent study 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 frequently used organisms for bioaccumulation monitoring at Greenland mine sites. These organisms have been assigned a key role because they are: 1) abundant at most Greenland mine sites; 2) stationary/sedentary; 3) effectively accumulate pollutants such as metals; 4) represent different trophic levels/habitats (i.e. have different food sources). Using monitoring organisms from different trophic levels/habitats allows 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, 2010b; Søndergaard, 2013a; Bach, 2020). After collection, the green-coloured growth tips of the seaweed have been 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). Further, the metal concentrations in the growth tips are considered to be a good proxy for the time-integrated dissolved metal concentration at the sampling site during the present growing season (spring-autumn) (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 l 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. Depuration of mussels in seawater to empty the gut content as recommended in some situations in ICES (2008) has not previously been part of the standard sampling procedure at mine sites in Greenland but is initiated at new sites. A sample size of 20 individuals pooled into one sample has been preferred for each size group. Also, the mean shell length (to the nearest 0.1 cm) of the mussel shells in each pooled sample is usually 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 was 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) undertook 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 nine 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). In addition to transplantation of blue mussels to the tidal zone at sampling sites, '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 typically 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). Further, 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 evaluated the 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). Lifespans of blue mussels in Greenland have been studied by Theisen (1973), who found that the lifespan of blue mussels from the Disko area on the Greenland West Coast ranged between 8 and 12 years for a 6 cm mussel, which is a common mussel size found at most sites in western Greenland and in south-eastern Greenland (in Northeast Greenland, blue mussels appear to be absent).

Sculpins (*Myoxocephalus* spp.) have been the preferred marine fish species for use 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, but shorthorn sculpin is 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 they are typically sampled by angling. As part of the sampling, fish length, fish weight, gender and liver weight have typically been determined. Most often, 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; Hansson et al., 2020) and blood (Hansson et al., 2020) 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.

*4.4. Assessment of 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 mine sites in Greenland has mainly focused on sampling of freshwater, lichens and Arctic char (*Salvelinus alpinus*, if present) (Søndergaard and Asmund, 2011; Bach, 2020). In addition, sampling of surface soil, dust, river sediment and spiders has been included in some of the previous studies (Søndergaard et al., 2012; Rambøll, 2013; Hansson et al., 2019).

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 (preferably 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 polyethylene/polypropylene syringe (without rubber o-ring) and 15 ml polyethylene vials. In addition to the water samples, in situ measurements of pH, temperature, electrical conductivity and redox potential/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 by filtering a large water sample (typically 1 l) 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 Ivittuut) as an 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 (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 the existence of 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, 2013, 2019; Bach, 2020). Transplanted lichens are placed on the ground on dead organic matter (i.e. not directly on the soil) and covered by a 1 × 1 cm mesh nylon net held in place by small flat pieces of rock. 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, previously metal accumulation by lichens could only be regarded as a relative (i.e. 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 study, a conversion factor was derived for (a rough) estimation of absolute deposition rates (at least for lead, zinc and cadmium) based on lichen transplants. By 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 atmospheric dust in different size fractions were initiated using a combined and portable optical particle counter and gravimetrical dust collector (DustTrak DRX). In this study, a field portable x-ray fluoresce spectrometer (pXRF, Olympus Vanta VMR) was tested and used for chemical characterisation of both natural and mine-related ground dust sources at Kangerlussuaq and at the anorthosite mine site at White Mountain (Søndergaard and Jørgensen, 2021).

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). Arctic char has been included in the monitoring program at Nalunaq due to the presence of an Arctic char population in the Kirkespir River near the mine (Bach, 2020). 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.

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 by hand using tweezers or captured in yellow cups containing water with a drop of liquid detergent. The cups were dug into the ground and placed at level with the ground surface. The collected spiders were preserved in ethanol prior to determination of species and chemical analyses.

### 4.5. Assessment of toxicological effects of pollutants

So far, most research and monitoring work conducted at Greenland mine sites has focused on assessing the dispersion and bioaccumulation of pollutants, while relatively few studies have assessed the direct toxicological effects of pollutants on organisms or changes in the presence/absence of organisms related to pollutants.

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 the foraminifer assemblage composition at polluted sites. In addition to these compositional changes, 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 sensitive and useful biomarker for evaluating trends in biological impacts from heavy metal pollution. Increased deformation of *Melonis barleeanus* during the mining period was observed in marine sediment cores as far as c. 10 km from the mine.

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 the mine closure was slow, and the impacted areas were 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 μg/g dry weight (d.w.), above which deterioration of faunal communities occurred. Above this threshold, diversity decreased dramatically, and dominance of sensitive and indifferent species was substituted by dominance of tolerant or opportunistic species. Benthic macrofauna changes were found in the Affarlikassaa Fjord and the inner Qaamarujuk Fjord within 2–4 km from the mine.

Sculpins are the most sedentary of the common fish species in Greenland 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. Sonne et al. (2014) undertook 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 40 km from the mine, respectively). Thus, the threshold values for Lowest Observed Effect Dose (LOED) were exceeded for the liver concentrations of mercury (reproduction), arsenic and cadmium (tissue lesions, biochemistry, growth and survival). No Observed Effect Dose (NOED) levels were exceeded for the liver concentrations of lead and zinc for 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 cofactor 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. The liver concentrations of mercury, arsenic, cadmium and lead exceeded the 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 (*Myoxocephlus 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 samples and analyses 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 a reduction of 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. *Orchomenella pinguis* was observed to effectively accumulate metals from both water and sediment, while simultaneously tolerating relatively high body concentrations of metals. Specifically, *Orchomenella pinguis* coped with high lead and zinc body concentrations without being severely affected 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 tropical species.

### *4.6. Sample preparation, chemical analyses and storage of samples and data*

Most analyses of environmental samples from mines in Greenland have been performed at the trace metal laboratory at DCE in Roskilde, Denmark accredited by the Danish Accreditation Fund (DANAK) following ISO 17025 for analyses of biota, sediment, fresh- and seawater for a range of elements. In addition to the trace element laboratory, a radioecology laboratory facility has recently been established, which will enable future radiological measurements. Methods and practices have evolved over the years following the technological advances in instrumentation etc. The following briefly describes the current methods and practices for sample preparation, chemical analyses and storage of samples and data.

For collection and storage until analyses, biota, sediment and soil samples are generally collected and stored in polyethylene bags and frozen at −20 °C until analyses. Water samples for element analyses are most often collected in polyethylene bottles or vials and stored cold at 5 °C or at room temperature. After reception of water samples in the laboratory, a clean nitric acid (i.e. Suprapure- or Ultrapure-grade) is added to keep the pH <2 (typically 1 ml/l for fresh water and 2 ml/l for seawater) and preserve the samples until analyses. An exception is water samples for low-level mercury analyses that are collected in borosilicate glass- or PTFE bottles and preserved with hydrochloric acid. A series of at least five blank samples is made for each batch of water samples using Milli-Q water and the same acid as for the samples.

Prior to the chemical analyses, sediment, soil and most biota samples are freeze-dried. Oven-drying is avoided to minimise the risk of mercury loss from the samples. After freeze drying, biota samples are most often homogenised to powder in an agate mortar to enable extraction of 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 d.w., which is the sample weight typically used for analyses. Samples of liver and muscle are typically not freeze-dried but cut directly out of the main sample after thawing, and the dry matter percentage is determined by oven-drying on a different subsample.

For most metal analyses of sediment, soil and biota, sample digestion is needed. The most frequently used digestion method involves a subsample (300 mg d.w. or 1 g wet weight biota and 200 mg d.w. sediment/soil) digested in half-concentrated Suprapure nitric acid in a microwave oven (currently an Anton Paar Multiwave 7000) according to Danish Standard, DS 259. For sediment and soil, sometimes a more destructive 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 element 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 (with element abbreviations): 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 the Agilent 7900 coupled to Agilent's Ultra High Matrix Introduction System and ISIS-3 Integrated Sample Introduction System has recently been developed for analyses of selected elements (V, Mn, Ni, Cu, Zn, Ag, Cd, Pb and U).

Additional available instruments and analyses include a direct mercury analyser (Milestone DMA-80), which enables analyses of mercury in sediment, soil and biota without prior sample digestion, an atomic fluorescence spectrometer (Millennium Merlin AFS) for low-level mercury analyses in water samples and a multicollector inductively coupled plasma mass spectrometer (MC-ICP-MS, Thermo Neptune XT) for precise isotope ratio analyses.

For data treatment, 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 three 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. 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.

For storage of samples, a large −20 °C freezer at DCE 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 prior to mining operations (baseline samples), and these 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 from 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 publicly available and used in the advisory work that DCE, in collaboration with GINR, provides to the Greenland authorities.

## 5. Lessons learned for future monitoring programs

Previous research and monitoring results have provided a good knowledge base for setting up adequate monitoring programs 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 the monitoring of freshwater for which pronounced temporal variations in water chemistry are typically observed, for instance due to spring flushes of pollutants caused by weathering of minerals during winter and release of accumulated weathering products (i.e. pollutants) released upon thawing. Also, seasonal stratigraphic mixing of seawater in the Arctic fjords can cause temporal variations in sea water chemistry, which should be taken into account in seawater monitoring.

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 chemically inert rock types such as feldspar and marble. Further, the monitoring program should be dynamic and subject to continuous revision according to its 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 the 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 measurements of pH, conductivity and total suspended solids); 2) bottom sediment from relevant lakes and marine sites; 3) lichens (preferably *Flavocetraria nivalis*); 4) seaweed (preferably *Fucus vesiculosus*); 5) mussels (preferably blue mussels, *Mytilus* spp.; typically 4–5 cm and 5–6 cm shell size intervals); 6) sculpins (*Myoxocephalus* spp.; liver, muscle and perhaps otoliths). If Arctic char are present, these should be included as well. Airborne particulates of selected particle size fractions (typically <2.5 μm and <10 μm) and total dust deposition may be added to the program. Sediment, lichens, seaweed, mussels and sculpins (and Arctic char) are usually sampled once a year, whereas more frequent water and dust sampling/characterisation may be required, especially during the operational phase of the project.

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. In case significant dispersion of pollutants is detected, the monitoring program should be adjusted and extended by using more comprehensive methods, and the scale and frequency of the monitoring should be adjusted accordingly to evaluate the dispersion and corresponding mitigation actions.

A combination of 'chemical/physical 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 the bioavailable pollutants of key monitoring organisms in the habitat. Also, annual sampling of, for example, growth tips of seaweed can 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 a high frequency of sampling (for instance daily/weekly/monthly) early in the project and possibly a subsequent reduction if no issues of concern appear.

## 6. Spatial and temporal trends of legacy pollution at Greenland mine sites

The cryolite mine at Ivittuut in South Greenland and the lead-zinc mines at Mestersvig and Maarmorilik in East and West Greenland are the most environmentally important mines in Greenland. At these older mine sites, of which Maarmorilik closed last in 1990, significant pollution of the environment was observed with mainly lead and zinc. This has given rise to frequent environmental assessments and numerous research studies since the environmental awareness started in the 1970s. For the other mines in Greenland's mining history, no significant pollution of the surrounding environment has been identified.

The cryolite mining at Ivittuut started in 1854, and environmental studies were not conducted until 1982, five years prior to the mine closure. The first studies identified significant pollution with lead and zinc in the surrounding Arsuk Fjord, and elevated concentrations of the two metals were measured in blue mussels (*Mytilus edulis*) and seaweed (*Fucus vesiculosus*) at distances up to c. 10-15 km from the mine during the first years of monitoring (Johansen et al., 2010b; Bach et al., 2014a). The pollution originated mainly from dissolution and transport of lead and zinc from waste rock deposited in the tidal zone along the coastline. Blue mussels near the mine showed elevated concentrations of lead, up to c. 1400 μg/g d.w. (typical background concentrations (bgc.) 0.5–5), and zinc, up to c. 370 μg/g d.w. (typical bgc. 80–150). Seaweed near the mine showed lead concentrations up to c. 150 μg/g d.w. (typical bgc. 0.1–0.5) and zinc concentrations up to c. 550 μg/g d.w. (typical bgc. 5–20). A decreasing trend of pollutants concentrations has been observed at Ivittuut since the first environmental study in 1982. During the period 1982–2013, lead concentrations in blue mussels and seaweed decreased by roughly a factor of 3, while zinc concentrations decreased by a slower rate of roughly a factor of 2. The area with elevated concentrations of lead and zinc at Ivittuut has also decreased in size and encompassed an area within roughly 5 km from the mine in 2013.

The lead-zinc mine at Mestersvig operated from 1956 to 1963, and the first environmental study was conducted in 1979. The environmental studies showed elevated concentrations of lead and zinc (and some cadmium and copper) in both in the marine and terrestrial environment surrounding the mine and harbour (Asmund et al., 1996; Johansen et al., 2008; Aastrup et al., 2018). Elevated concentrations were observed in a number of species including lichens (*Flavocetraria nivalis*), seaweed (*Fucus disticus*) and sculpins (*Myoxycephalus scorpius*) and in soil and river, beach and seafloor sediment. The maximum spatial extent of the pollution is unknown but assessed to at least 10–15 km from the mine and harbour. The main pollution sources were identified as a tailings deposit in Tunnel River just downslope from the mine entrance, situated c. 10 km inland from the harbour, dust dispersion of concentrate along the haul road from the mine to the harbour and spills of concentrate at Nyhavn (i.e. the harbour). Sometime between 1986 and 1991, part of the quay at Nyhavn collapsed, causing enhanced pollution in the harbour area in the subsequent years. Lichens contained lead concentrations up to 1530 μg/g d.w. (typical bgc. <1) and zinc concentrations up to 590 μg/g d.w. (typical bgc. <20). Seaweed contained lead concentrations up to 290 μg/g d.w. and zinc concentrations up to 400 μg/g d.w. Sculpins contained lead concentrations in the liver up to 4.1 μg/g d.w. (median for Nyhavn Bay; typical bgc. <0.1). A general trend toward decreasing concentrations of pollutants was observed at Mestersvig since the first study in 1979 but with a peak in 1991 near Nyhavn after the quay collapsed. During the period 1979–2014, the pollution at Nyhavn harbour decreased by roughly a factor of 3 for both lead and zinc as indicated by temporal trends in seaweed (Aastrup et al., 2018). During the last study in 2014, more than 50 years after the mine closure, elevated concentrations of lead and zinc were still observed near the mine, the haul road, Nyhavn and the adjacent fjord at a distance of at least 5 km away from the sources.

The lead-zinc mine at Maarmorilik started in 1973 and continued until 1990. The first environmental studies after the initiation of mining activities revealed pollution with mainly lead and zinc, not only in the small Affarlikassaa Fjord where tailings were disposed of but also in the adjacent Qaamarujuk Fjord system at a distance of at least 30 km from the mine. Dust dispersion led to elevated concentrations of lead and zinc in lichens, which could be measured up to a distance of 40 km from the mine. Elevated concentrations of other elements such as cadmium, mercury, copper and arsenic associated with the ore deposit were also identified (Elberling et al., 2002; Perner et al., 2010). During the mining period, the main pollution sources were identified as: 1) tailings discharged into the 2 $km^2$ sill fjord Affarlikassaa; 2) waste rock deposited on the mountain slopes and in the fjords; 3) dispersion of metal-laden dust from ore extraction, transport, processing, loading of concentrate etc. Elevated lead and zinc concentrations were observed in seawater, sediments and a long range of marine organisms as well as in soil, lichens and spiders in the terrestrial environment (Larsen et al., 2001; Elberling et al., 2002; Johansen et al., 2006; Hansson et al., 2019). Dissolved lead concentrations in bottom water in the Affarlikassaa Fjord up to 440 μg/l (typical bgc. 0.01–0.03) and zinc concentrations up to 790 μg/l (typical bgc. 0.3–0.8) were measured. The peak concentrations measured in key monitoring species were as follows (Johansen et al., 1997): Blue mussels (*Mytilus* spp.) showed lead concentrations up to 3850 μg/g d.w. and zinc concentrations up to 1330 μg/g d.w. Seaweed (*Fucus vesiculosus*) displayed lead concentrations up to 166 μg/g d.w. and zinc concentrations up to 520 μg/g d.w. After the mine closure in 1990, the pollution decreased markedly, and the tailings in the bottom of the Affarlikassaa Fjord gradually became covered with natural sediments and were no longer considered the main pollution source. The concentrations decreased by roughly a factor of 10 for lead and roughly a factor of 2–3 for zinc as indicated by the concentrations in blue mussels and seaweed at polluted sites recorded near the mine (Schiedek et al., 2009). Since around year 2000, the concentrations of pollutants in the area have remained relatively stable with no clear decreasing trend. During the last study in 2017, the Qaamarujuk Fjord was still polluted with mainly lead and zinc, and elevated lead and zinc concentrations were observed in surface sediment and seaweed 12 km from the mine (Hansson et al., 2020). Similarly, dust dispersion and deposition with mainly lead, zinc and cadmium were still significant, and elevated concentrations were measured in surface soil and lichens 12 km from the mine (Hansson et al., 2019). The continuous pollution after closure of the Maarmorilik mine is considered mainly due to continuous degradation of waste rock left at the mountainsides exposed to weathering processes, leaching and wind transport as well as dust dispersion of residues of ore and concentrate still left in the mining town area. The waste rock is situated on steep slopes extending several hundred meters above sea level up to the mine entrances at approximately 600 m altitude, and the frequent strong winds from the Greenland Ice Sheet promote wind transportation.

In conclusion, three to five decades after the mine closures at Ivittuut, Mestersvig and Maarmorilik, the pollution has decreased, but the mining activities have left a legacy of pollution extending at least 5–12 km away from the sources. The pollution is likely to continue for many decades to come due to weathering of waste materials left for erosion and transport, and these are difficult or impossible to retrieve and cover. Moreover, climate change may alter the pollution in the areas since tailings and waste rock now frozen and part of the permafrost may thaw and become exposed to weathering and pollutant transport. This is particularly the case for the waste rock dumps at Maarmorilik and the tailings deposit close to Tunnel River at Mestersvig, emphasising the need for continuous environmental studies at these old legacy mine sites.

## 7. The role of environmental research and monitoring, regulations and EIAs in minimising the environmental impact of mine sites in Greenland and the lessons learned

On the road 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 concentrations and effects. A diverse toolbox of monitoring methods is needed to be able to select the most efficient methods for determining 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) to be applied when awarding mining licenses. Furthermore, monitoring results are 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. This has likely reduced the overall environmental impact at Maarmorilik significantly in the long term. 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 issue was identified by the monitoring program during the mine operation. In consequence, the authorities demanded initiation of mitigation actions, and a dust suppressant was subsequently applied to the roads. This led to reduced dust dispersion (Søndergaard and Asmund, 2011), and no significant dust dispersion has been observed in the surrounding area at Seqi after the mine closure (Søndergaard, 2019).

Also, various lessons have been learned with regard to minimising the environmental impact at Greenland mine sites based on the results of the past research and monitoring. These have been applied in other projects and include, among others, 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 these results and site-specific conditions into account when depositing the waste. Specifically, waste containing elements and minerals of environmental concern, as identified by the leach testing, should never be deposited at exposed mountain slopes or in the sea, where it is difficult or impossible to access, control and retrieve (as in Maarmorilik) or in the tidal zone, where it is exposed to high rates of leaching and transport (as in Ivittuut). Tailings should be deposited following the principles of Best Available Technology/Best Environmental Practice, and site- and mine-specific factors must be considered, implying that tailings should never be deposited in riverbeds exposed to erosion, water and wind transport (as in Mestersvig). In areas with permafrost, waste materials are likely to be very difficult to retrieve if they become frozen and may need blasting to be removed with the associated risk of dust dispersion of pollutants. Therefore, it is critical to adopt an adequate long-term solution in these areas before the disposal. Furthermore, the ongoing climate change affecting temperatures and precipitation must be considered for long-term storage. Climate change is a significant unknown regarding the tailings deposit near Tunnel River at Mestersvig and the waste rock deposit in Maarmorilik, where a significant amount of material is now frozen but may thaw in the future and thus become exposed to weathering, leaching and transport, potentially augmenting the pollution here.

The experiences gained, together with knowledge obtained from mines in other parts of the world, are now part of the knowledge base at DCE and GINR and form the basis for the advisory work provided to the Greenland authorities on mining projects. This advisory work includes evaluation of EIAs, 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 the significant mining operations at Mestersvig in the 1950–60s and Maarmorilik in the 1970–80s (Table 1). At these three mine sites, none or very few environmental studies were conducted prior to the mining activities. 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 has been identified in the areas of Nalunaq, Seqi, Aappaluttoq and White Mountain, where mining was/is undertaken in the 2000–2020s. 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 licenses, based on the highest international standards and the knowledge obtained from legacy mines in Greenland and elsewhere.

## 8. Future perspectives and final remarks

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 (disturbance from increased shipping, helicopter transport etc.) in the areas of concern. Furthermore, there is a potential to involve the local community near a mine site in the monitoring to take advantage of local ecological knowledge, 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 that 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 and for minimising the environmental impact of mining. 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 mine sites. Thus, 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, and the adapted monitoring programs should include a diverse suite of techniques including both chemical/physical and biological tools. Programs should also take temporal variations in the discharge of pollutants unique to the Arctic into account, such as spring flushes of winter-generated pollutants or seasonal hydrographical processes in the fjords. Owing to the continued research in mining areas and the development of new techniques, the future environmental monitoring of mining activities in Greenland is likely to encompass a range of additional tools for assessing environmental impacts. Future monitoring programs may also include results from regional monitoring programs and assessment of cumulative effects of mining activities on a larger scale. The current climate change with increasing temperatures in the Arctic calls for continuing the monitoring of legacy mine sites due to thawing of previously frozen material, increasing weathering rates and changes in the hydrological regime that may affect the release of pollutants.

## CRediT authorship contribution statement

**Jens Søndergaard:** Conceptualization, Methodology, Investigation, Visualization, Writing – original draft. **Anders Mosbech:** Conceptualization, Writing – review & editing, Funding acquisition.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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