# Lancaster Environment Centre Lancaster University

# Investigating the sources, transfer, and fate of microplastics in the Arctic marine environment

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This thesis is submitted in partial fulfilment of the requirements for the degree of Doctor of Philosophy

September 2024



#### **Abstract**

Contamination of the world's oceans by plastic waste continues to be a widespread global concern, with microplastics posing a particular threat due to their ability to be transported globally and ingested by a wider range of marine biota. In the Arctic, microplastics have now been reported in sea ice, snow, surface and sub-surface waters and deep-sea sediment, as well as Arctic biota, but large parts of the Arctic have not been subject to monitoring. In addition, scientific appraisals into the sources, cycling and fate of microplastics reveal multiple knowledge gaps, including the effects of these pollutants on Arctic ecosystems. This thesis provides new insights into the occurrence of microplastics particles in the Beaufort Sea where particles were found at much higher concentrations than other areas of the Arctic. These high concentrations may also be found in sea ice, with results showing sea ice formation led to the incorporation of microplastic particles from seawater with very high concentrations in the uppermost layer of ice (ice - atmosphere interface), reinforcing the notion that sea ice can act as a significant 'temporary' sink for microplastic pollution. This raises concerns for Arctic biota, however results showed zooplankton species C. finmarchicus contained very low microplastic concentrations, suggesting they are unlikely to be acting as a transfer mechanism for microplastics from surface waters to the deep ocean. Overall, the results from this study suggest future research should focus on assessing potential sinks for microplastic pollution in the Arctic, as well as the effects of microplastic ingestion in sympagic (ice-associated) organisms.

#### **Acknowledgments**

I would like to begin by expressing my deepest gratitude to my supervisory team. To Crispin Halsall, thank you for your unwavering guidance and for encouraging me to keep going through the challenges. To Alice Horton, thank you for inspiring me to pursue this PhD in the first place, and for your continued support and mentorship throughout. To Jan Kaiser, I truly appreciate your quick responses and meticulous attention to detail, which have been invaluable. And to Yueng-Djern Lenn, your positivity and constant encouragement have been a great source of motivation and kept my spirits high. I am immensely grateful for the collective knowledge and experience you all have shared with me during this process.

I would also like to thank Envision DTP, and especially the friends I have made through the program. Catching up at the yearly conferences has been a highlight of this journey.

To my family and friends, your endless support has carried me to this point. Your words of encouragement have meant the world. A special thank you to my mum, for making me believe I can achieve anything.

Finally, to my husband Josh, planning a wedding while completing a PhD was no small feat, but your love and support have made it all possible. You are my rock, and I couldn't have done this without you by my side.

"Perseverance is not a long race; it is many short races one after the other." – Walter Elliot

### **Table of Contents**

Αŀ	bstract	2
Αd	cknowledgments	3
In	troduction	8
Ba	ackground and rationale	8
Re	esearch aims and objectives	9
O۱	verview of thesis	10
Re	eferences	12
1.	Plastic pollution in the Arctic marine environment	13
1.	1 Introduction	13
1.	2 Sources	16
	1.2.1 Marine inputs	17
	1.2.2 Terrestrial sources.	17
1.	3 Transport	18
	1.3.1 Ocean transport	19
	1.3.2 River transport.	20
	1.3.3 Sea ice transport.	22
	1.3.4 Bio-transport	22
	1.3.5 Atmospheric transport	23
1.	4 Fate	24
	1.4.1 Sea ice	25
	1.4.2 Surface and sub-surface waters	28
	1.4.3 Deep-sea sediments	29
1.	5 Summary	30
Re	eferences	32
2.	The Beaufort Sea: an accumulation zone for microplastic pollution	38
Αŀ	bstract	38
2.	1 Introduction	38
2.	2 Materials & methods	41
	2.2.1 Sample collection.	41
	2.2.2 Sample processing and analysis	42

2.2.3 Statistical Analysis.	43
2.2.4 Contamination mitigation and blanks	43
2.3 Results	44
2.3.1 Quality controls.	44
2.3.2 Microplastics in seawater	44
2.3.3 Microplastic polymer types	45
2.3.4 Microplastic size	46
2.3.5 Temperature and salinity	47
2.4 Discussion	48
2.4.1 Microplastics in the Beaufort Sea.	48
2.4.2 Microplastics in the water column	50
2.4.2.1 Microplastics in surface samples.	51
2.4.2.2 Microplastics in subsurface samples	51
2.4.3 Microplastic polymer types and size	53
2.5 Conclusion	54
Acknowledgements	55
References	55
Supporting Information	60
3. Mechanistic understanding of microplastic fate in sea ice: a simulated freeze –	thaw
study	63
Abstract	63
3.1 Introduction	63
3.2 Materials and Methods	66
3.2.1 Experimental setup.	66
3.2.2 Quality controls and blanks	67
3.2.3 Experimental procedure and sampling protocol	67
3.2.4 Sample processing and analysis	70
3.2.5 Particle size and density.	70
3.2.6 Microplastic quantification.	71
3.2.7 Enrichment Factor (EF) for microplastics in sea ice	71
3.3 Results	71
3.3.1 Quality controls and accounting for spike particles in the sea ice chamber	71

	3.3.2 Visual observations of particle size and density in seawater	/2
	3.3.3 Observations of particle behaviour in seawater	73
	3.3.4 Observations of particle behaviour in sea ice	75
	3.3.5 Observations of particle behaviour in meltwater	76
3.	4 Discussion	77
	3.4.1 Observations of particle behaviour in seawater	78
	3.4.2 Observations of particle behaviour in ice.	79
	3.4.3 Observations of particle behaviour in meltwater	81
3.	5 Conclusion	82
Αc	cknowledgements	82
Re	eferences	82
Su	upporting Information	86
4.	Measuring the body burden of microplastics in Arctic Copepods from 90	the Fram Strait.
ΑŁ	bstract	90
4.	1 Introduction	91
4.	2 Materials & methods	94
	4.2.1 Sample collection.	94
	4.2.2 Sample processing	95
	4.2.2.1 Picking	95
	4.2.2.2 Digestion.	95
	4.2.3 Microplastic identification	96
	4.2.4 Contamination mitigation and blanks	96
4.	3 Results	97
	4.3.1 Microplastic findings in C. finmarchicus.	97
	4.3.2 Microplastic polymer types	97
	4.3.3 Microplastic particle size	98
4.	4 Discussion	99
	4.4.1 Microplastic concentrations in C. finmarchicus.	100
	4.4.1.2 Seasonal variations in microplastic concentrations	101
	4.4.2 Microplastic polymer types and size	102
4	5 Conclusion	104

Acknowledgements	104
References	104
Supporting Information	109
5. Conclusions and areas of further research	111
5.1 Summary of results.	111
5.2 Areas for further research	115

#### Introduction

#### **Background and rationale**

The production and consumption of plastic has been growing at an exponential rate since mass manufacturing began in the 1940s (Wilcox et al., 2019). In 2020, global plastic production was around 450 million, a huge increase from the 2 million tons produced in the 1950s (EEA, 2024). Geyer et al (2017) estimated that 2,500 million tonnes of plastics, or 30% of all plastics ever produced, are currently in use. Between 1950 and 2015, cumulative waste generation of primary and secondary (recycled) plastic waste amounted to 6,300 million tonnes. Of this, approximately 12% of plastics have been incinerated and 9% have been recycled. Around 60% of all plastics ever produced were discarded and are accumulating in landfills or in the natural environment. None of the commonly used plastics are biodegradable, and most of the monomers used to make plastics, including ethylene and propylene, are derived from fossil hydrocarbons (Geyer et al., 2017). This has subsequently led to a significant increase in plastic waste accumulating in the environment. The potential impacts of global plastic pollution are extensive, having both geophysical and biological impacts, and could put added pressure on already vulnerable ecosystems (Macleod et al., 2021). There have been numerous studies showing these impacts, including entanglement of organisms in plastic debris and ingestion of plastics, which has been shown to cause various health issues.

It is estimated that 19 to 23 million metric tons of plastic waste generated globally in 2016 entered the marine environment, and this figure is estimated to reach up to 53 million metric tons per year by 2030 (Borrelle et al., 2020). Once in the marine environment, plastic debris is exposed to a number of physical, chemical and biological processes which result in the breaking down of larger plastic pieces into much smaller pieces; known as microplastics (Andrady, 2011). Microplastics, typically defined as plastic particles ranging in diameter from 1 µm to 5 mm (Frias and Nash, 2019), can be characterised as primary or secondary (Cole et al., 2011). Primary microplastics are manufactured to be of microscopic size for uses in cosmetics and medicine, whereas secondary microplastics are derived from the breakdown of larger plastic debris. Microplastics can also act as both a direct (particulate) hazard and an indirect (chemical) hazard to a range of marine organisms (Horton et al., 2017).

Despite the production of plastic taking place in highly urbanised, industrial areas of the world, microplastic pollution has been reported in even the most remote environments, far from human activities. However, there are currently large knowledge gaps regarding the key pathways of these contaminants to remote areas, and their fate within these environments. Microplastics in the Arctic have entered terrestrial and marine systems, as well as the cryosphere and the atmosphere, through a number of transport pathways including ocean currents and atmospheric inputs. The origins of these microplastics are thought to stem from long-range sources, although some pollution is from local sources such as mismanaged waste and fisheries. The Arctic Ocean's currents and water masses are influenced by waters from the North Atlantic and North Pacific, as well as river inputs. Because of this, it is thought that the Arctic could be acting as an accumulation zone or dead end for microplastic pollution. There is also evidence to suggest the spread of plastic pollution will be exacerbated by climate change (Ford et al., 2022). Global warming has led to mass loss from ice sheets and glaciers, reductions in snow cover and sea ice extent and thickness. A 90% reduction in multi-year ice has been observed between 1979 and 2018, suggesting a transition to younger ice. Warming in the Arctic has been amplified by feedbacks from the loss of summer sea ice and spring snow cover on land, and surface air temperature has increased by more than double the global average over the last two decades (IPCC, 2019). Sea ice has been identified as a significant sink for microplastic pollution, and this advanced melting could enhance the release of microplastics (Peeken et al., 2018). Microplastics pose a threat to a number of Arctic species, from mammals to smaller multicellular organisms that ingest them, such as copepods. These organisms are abundant in the sea ice zone, either living within the brine channel system of the ice-crystal matrix or inhabiting the ice-water interface (Ásmundsdóttir and Schulz, 2020). However, the effects microplastics have on these organisms is largely unknown.

#### Research aims and objectives

The overarching aims of this thesis was to better understand the occurrence, transfer and fate of microplastics in surface marine waters of the Arctic. Specifically, the study involved the following objectives:

• Identify knowledge gaps concerning the sources, occurrence, and ecological impacts of microplastics in the Arctic marine environment.

- Quantify microplastic abundance in Arctic seawater and compare concentrations by depth.
- Examine the uptake and enrichment of microplastics by artificial sea ice in a controlled laboratory setting during ice growth and melt.
- Explore the transfer of microplastics from the abiotic to the biotic environment via the base of the Arctic marine food web.

#### **Overview of thesis**

This thesis aims to explore the dynamics of microplastic pollution in the Arctic, through four distinct chapters that explore the transfer and fate of microplastics in both biotic and abiotic samples. **Chapter 1** serves as a comprehensive review of the existing literature on plastic pollution in the Arctic Ocean, synthesizing decades of research to provide a foundational understanding of the scope and scale of this pervasive environmental issue. This chapter has been published in the book titled Plastic Pollution in the Global Ocean by Alice Horton and serves to provide background detail and context for the rest of the thesis.

The relationship between microplastic abundance and depth is explored in **Chapter 2**. This chapter details the results of a study investigating the Beaufort Sea as a potential accumulation zone for microplastic pollution. Sampling was conducted onboard the Sikuliak cruise during September 2018 in collaboration with the Changing Arctic Oceans PEANUTS project. Water samples were taken from two distinct layers of the water column at multiple locations in the Beaufort Sea, to determine whether microplastic number concentrations were high, and whether these concentrations differed with depth.

Chapter 3 provides findings from a simulated freeze up – thaw down experiment, conducted in the Roland von Glasow Air-Sea-Ice Chamber (RvG-ASIC) at the University of East Anglia, Norwich, UK, to investigate the mechanisms of microplastic uptake within sea ice. This state-of-the-art facility was built for the purpose of studying environmental processes in polar regions, highlighting processes by which these pollutants become entrapped and transported within the ice matrix. This facility allows users to simulate sea ice growth and melt in a controlled environment. Microplastics were 'spiked' into the sea ice chamber and samples from surface and sub-surface seawater, ice, and meltwater were taken to establish

microplastic number concentrations. The microplastic spikes used in Chapter 3 were polyethylene, chosen due to the prevalence of polyethylene in environmental samples and ease of identification A range of sizes and densities were used in an attempt to create a more realistic study.

Chapter 4 considers the presence of microplastics in Arctic organisms. Zooplankton samples were collected during two cruises on the RRS James Clark Ross: May 2018 (JR17005) and August 2019 (JR18007). The focus of the cruises was the Fram Strait, where deep basins coincide with seasonal sea ice, creating ideal conditions for Calanus to accumulate large fat stores and enter diapause in deep ocean layers, where they may inadvertently transport microplastics from surface waters to the deep ocean. *C. finmarchicus* in stage C5 were chosen to investigate the potential of these organisms to transport microplastics from surface waters to the deep ocean during their seasonal migration. This work was a collaborative effort with researchers at NOC Southampton.

Finally, **Chapter 5** draws together the findings discussed in this thesis and identifies areas of potential future research to build upon observations from this study.

All experimental design, development of methods, data generation, data analysis, and interpretation presented in this thesis were carried out by the author. For chapter 2, sample collection was carried out as part of the Changing Arctic Oceans project and not by the author, although samples were collected with the intention of microplastic analysis. For chapter 4, samples were collected as part of two research cruises on the RRS James Clark Ross: May 2018 (JR17005) and August 2019 (JR18007), and not by the author. The author was responsible for the planning and execution of all laboratory activities, as well as the statistical analysis and visualisation of results. The thesis was written entirely by the author. Supervisors provided guidance and advice throughout the project, including feedback on experimental design, discussion of data interpretation, and comments on draft chapters. No other individuals contributed directly to the experimental or analytical work contained within this thesis.

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## 1. Plastic pollution in the Arctic marine environment

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#### 1.1 Introduction

Concern about pollution in the Arctic and growing environmental pressures across this region gave rise to the Arctic Environmental Protection Strategy in 1991; a circumpolar agreement focused on environmental monitoring, protection of the Arctic environment and later, environmental sustainability (see UNEP). A number of working groups with specific environmental remits were created that operate under the Arctic Council. One of these groups, the Arctic Monitoring Assessment Programme (AMAP) reports widely on pollutants like radioactive substances, mercury/heavy metals, and persistent organic pollutants, particularly with regards to human health and ecosystem exposure (AMAP, 2023). A growing area of concern is marine litter and microplastics, and a programme to assess these contaminants and their impact on food-webs is underway (AMAP, 2023). This has included an extensive assessment of marine plastic litter coordinated through the Protection of the Arctic Marine Environment (PAME), another Arctic Council working group whose remit covers the protection and sustainable use of the marine environment.

Polar Regions are increasingly receiving, and impacted by plastic waste (Horton and Barnes, 2020). Arctic beaches are visibly polluted with plastic debris, and the rates of this pollution are continuing to rise as Arctic populations grow and tourism increases (Polasek et al., 2017). Tourism has been identified as an important source of litter in temperate coastal regions, and it is suspected that it will soon have a large contribution to plastic pollution in the Arctic (Tekman et al., 2017). Polasek et al (2017) conducted a project to remove and document marine debris from five coastal National Park Service units in Alaska. The remote, relatively pristine nature of the Alaskan coastline was found to have a surprisingly high accumulation of anthropogenically-derived debris. Approximately 80 km of coastline were cleaned, and over 10,000 kg of marine debris was collected. Out of the 28 beaches surveyed, hard plastics were found at every one, and ropes/netting were found at almost all beaches. Overall, plastic

contributed to 60% of the total weight of debris. The results suggest local geography and substrate of a beach likely has a strong effect on the amount and concentration of debris collected. There is a significant lack of information regarding the concentration, distribution, and types of plastic debris on Arctic beaches, especially those in more remote higher-latitude locations (Bergmann et al., 2017).

The issue of macroplastic pollution is one that we can visually acknowledge, however a growing concern comes from plastic particles that can be difficult to observe e.g., microplastics ( $\leq 5$  mm in diameter) and nanoplastics ( $\leq 1$   $\mu$ m in diameter). Currently, microplastics in the Arctic have been observed in sea ice (Obbard et al., 2014, Bergmann et al., 2016, Peeken et al., 2018), snow (Bergmann et al., 2019), surface and sub-surface waters (Lusher et al., 2015, Kanhai et al., 2018) and deep-sea sediment (Bergmann et al., 2017), as well as Arctic biota (Provencher et al., 2010, Kühn et al., 2018, Morgana et al., 2018). A range of polymers have been identified in samples suggesting multiple sources, both local and longrange, with many particles being in the lower size ranges (<500 μm). These smaller particles are more readily available to marine biota (Lusher et al., 2015), although very few studies have been carried out on the effects of plastic ingestion on Arctic marine organisms. It is suggested that ingestion of smaller plastic particles, which can carry harmful toxins both on the surface and within the polymer matrix, can affect the mobility, reproduction and general functioning of the organisms that consume them (Kühn et al., 2015). The ingestion of microplastics by blue mussels (Mytilus edulis) (Sundet et al., 2016) and snow crabs (Chionoecetes opilio) (Sundet et al., 2014) was investigated in Svalbard, with results showing that both species ingested microplastics, with 90% occurrence and an average of 9.5 items per individual for the mussels and 20% incidence for the snow crabs (PAME, 2019). Trevail et al (2015) explain that plastic ingestion levels in the Arctic are generally minor compared to levels at lower latitudes. However, levels of ingestion are not as low as would be expected given the remoteness of some of the study regions. This raises questions about the sources, transport, and fate of plastic debris and microplastics in the Arctic as a whole.

Microplastics have been discovered in both surface and sub-surface marine waters, demonstrating that they are readily available for incorporation into sea ice (Lusher et al., 2015), which has been identified as a sink for microplastic particles. Currently, there's a lack of knowledge concerning the processes and rates of microplastic incorporation within sea ice,

and their possible effect on sea ice properties, such as influencing the albedo (proportion of incident sunlight reflected) of surface snow (Geilfus et al., 2019). The Arctic is currently seeing record lows in sea ice extent, which could affect plastic particle dynamics and transport across the Arctic Ocean and see the release of large amounts of legacy microplastics that have previously been entrained in the ice (Obbard et al., 2014). Arctic sea ice has been recognised as a repository for organic contaminants present in seawater but also deposited from the atmosphere (e.g. Pucko et al., 2015, Garnett et al., 2021) and microplastics have also been observed in sea ice and the ice-rafted snowpack (Peeken et al., 2018). As sea ice forms it will incorporate particles that are suspended in the water column, which will then be transported long distances as sea ice drifts. Contaminants will then be deposited when the sea ice melts, making them available to sympagic (ice-associated) and pelagic marine organisms (Obbard, 2018).

Research into plastic pollution in polar regions is lacking, especially regarding the fate of macroplastic litter and the formation and/or transport of microplastic particles. Studies have previously focussed on identifying the abundance, sources, and accumulation patterns of microplastics, but the effects that plastic pollution is having on Arctic ecosystems is unknown (Horton and Barnes, 2020). Tirelli et al (2020) explain that the first records of plastic pollution in the Arctic date back to the early 1960s, when the ingestion of plastic fragments was reported in Canadian seabirds (Threlfall, 1968, Provencher et al., 2015) and more thoroughly in the 1970s, when the total amount of beached macro-litter items in the Bering Sea increased from over 2200 to more than 5300 in only 2 years – 1972 to 1974 (Merrell, 1980). Research on marine mammals and entanglement reached a peak during the 1980s and 1990s in Arctic waters of Alaska and adjacent areas (Bering Sea and Gulf of Alaska) but monitoring has since reduced, and in the rest of the Arctic, knowledge is fragmented and covers only some groups or species (PAME, 2019). The current knowledge on plastic pollution in the Arctic marine environment relates to areas where human activities are concentrated, such as the Barents Sea, Norwegian Sea, and Bering Sea, with few data available for the Central Arctic Ocean and the coastal areas around it in Siberia, Arctic Alaska, mainland Canada, and the Canadian Arctic Archipelago (PAME, 2019). These regions are clearly more difficult to access, hence the lack of data, but if we are to get a homogenous understanding of marine plastic pollution in the Arctic, it is vital we include these areas. Against this backdrop of pollution pressures, the Arctic

is warming on average more than twice the global mean (Jansen et al., 2020). From 1971-2019, the annually averaged Arctic near-surface air temperature increased by 3.1°C, three times faster than the global average (AMAP, 2021), further demonstrating the importance of understanding the fate of plastic pollution in this rapidly changing environment. This rapid warming is leading to advanced thinning and melting of sea ice with implications for the global climate system. According to the US National Snow & Ice Data Center (NSIDC) and reported by AMAP, Arctic sea ice extent in September (the month with lowest sea ice cover) has declined by 43% between 1979 and 2019, and most areas are seeing a decline not only during the summer months but all year round. It is also reported that all Arctic regions are facing net loss of land ice, with Greenland accounting for 51% of the total. These losses of land ice in the Arctic are a significant contributor to global sea level rise (IPCC Special Report on the Ocean and Cryosphere in a Changing Climate). Arctic rivers are also experiencing shorter freezing periods, and ice thickness in the northernmost Arctic rivers is decreasing. The volume of water flowing through these rivers and into the Arctic Ocean has increased by 7.8% over the last 5 decades (AMAP, 2021) and has the potential to alter pollutant transport pathways to Arctic coastal seas and the wider Arctic Ocean. Increased anthropogenic pollution has the potential to exacerbate the negative effects of climate change on ecosystems (e.g., melting sea ice releasing large concentrations of microplastics back into the environment), and vice versa, for example, increased extreme weather as a result of climate change has the potential to worsen the spread of plastic pollution in the environment (Ford et al., 2021).

#### 1.2 Sources

Due to its low-density, plastic debris can be transported long distances, meaning areas where the debris deposits are most likely not where the source of pollution is found. Sources of plastic pollution to the Arctic can be split into either **terrestrial** or **marine-based** origins, for example, discarded fishing gear has been highlighted as one of the main contributors to floating plastic debris in the Arctic (PAME, 2019), which originates in the marine environment from both local and long-range sources. The remote nature of the Arctic would suggest that most plastic debris entering the region is from long-range sources, travelling via ocean currents, atmospheric pathways, or riverine inputs.

1.2.1 Marine inputs. The main sectors of maritime activity in the Arctic include fishing, both commercial and recreational, aquaculture, and shipping, including cruise tourism (PAME, 2019). Plastic debris associated with these activities such as nets, floats and rope can enter the marine environment through accidental loss, poor waste management or illegal dumping (Sheavly and Register, 2007). These macroplastics create microplastics through fragmentation, by several processes including photolysis, thermal-oxidation, hydrolysis, and physical abrasion (Browne, 2015).

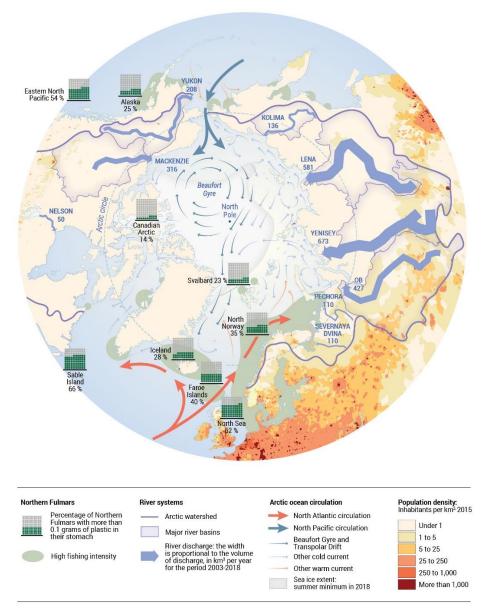
Bergmann et al (2017) conducted a citizen science experiment which involved participants of a tourist cruise documenting and collecting plastic debris on Arctic beaches. Over the six beaches surveyed, a total of 991 kg of plastic debris was collected, with the highest quantities found on Reinstrandodden in Svalbard (524 g m<sup>-2</sup>). The Svalbard beaches showed particularly high concentrations of fisheries-related plastic, with such items including ropes, buoys, floats, nets, and smaller broken pieces of the afore mentioned. These items accounted for 44-100 % of the litter mass, with other plastic items such as packaging material, bottles, toys, and other single-use plastics also present. Bergmann et al (2017) explain that the dominance of fisheries-related litter on the beaches of Svalbard may be partly due to the unit used to express litter quantities (g m<sup>-2</sup>), which is sensitive to the occurrence of single, particularly heavy items such as fishing nets at Reinstrandodden, highlighting the importance of transparency when reporting methodologies. However, its strong dominance at all beaches examined clearly points to fisheries as a major source of litter pollution. Most fishing gear that ends up on beaches and can be analysed is made of material that floats, meaning material that does not float, such as set nets or crab pots, do not end up on beaches and remain in the sea (PAME, 2019). During the citizen science experiment, participants took images to demonstrate the effects of plastic litter on Arctic biota, which mostly portrayed wildlife entangled in fishing nets and ropes. Several Arctic species were affected, including Arctic whitlow-grass (Draba bellii), seabirds (specifically Sterna paradisaea), and terrestrial and marine mammals such as harbour seals (*Phoca vitulina*) and polar bears (*Ursus maritimus*).

1.2.2 Terrestrial sources. Plastic litter concentrations from local land sources should be relatively low in the Arctic due to its remote nature and relatively low population of 4 million, which are primarily concentrated in townships and capital cities, leaving the central Arctic area with a comparatively low population (Heleniak and Bogoyavlensky, 2015). However,

increased discharge from the major Arctic rivers (North American and Eurasian rivers) that have very large catchments extending to sub-Arctic regions may be a significant and growing source of plastic debris to the coastal Arctic Seas. The remoteness of the central Arctic and the sparse coastal-based communities is unlikely to give rise to plastic waste from terrestrial sources on an Arctic-wide basis. Granberg et al (2017) explain that the release of untreated sewage into coastal environments is one of the most significant unregulated local pollution sources in the Arctic, where sewage treatment is generally lacking, resulting in a continuous discharge of sewage and wastewater from households and commercial industries directly to coastal waters (PAME, 2019, Granberg et al., 2017). Household waste is likely to contain microplastics, primarily fibres, originating from cosmetics and washing synthetic polymer clothing (Napper and Thompson, 2016). Kirkelund et al (2017) found that in small Arctic communities, the traditional waste management solutions are uncontrolled waste dumps and simple incinerators with no or limited flue gas treatment. Magnusson et al (2016) investigated wastewater outputs in various Nordic Sewage Treatment Plants (STPs) and found that those plants using only mechanical treatments such as those in Iceland had limited particle retention, and outputs of microplastics amounted to ~6,350,000 microplastics (of diameters ≥300 µm) per hour from Klettagarðar and ~2,230,000 microplastics per hour from Hafnarfjörður. An increase in tourism combined with rudimentary waste management, will inevitably lead to higher levels of plastic pollution from local sources in the vicinity of Arctic communities and nearby coastal regions.

#### 1.3 Transport

Contaminants such as macroplastic litter and microplastics can enter the Arctic via a combination of processes, such as surface oceanic currents, freshwater input from rivers, and atmospheric processes, which all transport contaminants from long-range sources. Zarfl and Matthies (2010) considered the possibility that marine plastic particles could act as transport vectors for organic pollutants to the Arctic. The study used thermodynamically based model calculations to determine the relative importance of plastic debris as a source of persistent bioaccumulative toxins (PBTs) to the remote Arctic Ocean. Their models conclude that transport via atmospheric and ocean currents are orders of magnitude larger than via plastic particles, determining that the contribution of PBTs from plastic debris may be small compared with annual PBT flux from other global transport mechanisms.



**Figure 1.** Plastic input into the Arctic Ocean highlighting Arctic Ocean circulation, sea ice extent, major river basins, river basin water discharges to the Arctic coastal seas, and plastic ingestion by Northern Fulmars. Source: GRID-Arendal 2019, <a href="https://www.grida.no/resources/13350">https://www.grida.no/resources/13350</a>. Creators; Philippe Rekacewicz, Riccardo Pravettoni, and Nieves Lopez Izquierdo.

1.3.1 Ocean transport. From Figure 1, the central Arctic Ocean is surrounded by shallow coastal seas as well as passages and straits: the Barents Sea, Greenland Sea, Norwegian Sea, and the Fram Strait, located in the European region of the Arctic. The Labrador Sea, Davis

Strait, Baffin Bay, the Northern Passages, Hudson Strait and Hudson Bay, and the Beaufort Sea, all of which are located around Canada. The Chukchi Sea and Bering Sea surround Alaska, and the East Siberian Sea, Laptev Sea and Kara Sea around northern Russia (PAME, 2013). Waters flowing into the Arctic basin include Atlantic waters entering via the Fram Strait as well as into the Barents Sea from the Norwegian Sea, and Pacific waters flowing from the Bering Sea via the Bering Strait and across the Chukchi Sea (Loeng et al., 2005, Tirelli et al., 2020).

Kanhai et al (2018) report that the oceanic currents which supply the greatest water volumes to the Arctic Ocean, including The West Spitsbergen Current, a branch of the North Atlantic current which flows northwards through the eastern side of the Fram Strait, and the Bering Sea current bringing Pacific Ocean water into the Arctic and flows along the Siberian coastline, may also transport plastics, with the estimated flux ranging between 62,000 to 105,000 tonnes per year (Zarfl and Matthies, 2010). Onink et al (2019) studied the role of various physical processes, such as wind-driven Ekman currents, geostrophic currents, and waveinduced Stokes drift on the global surface distribution of floating microplastics. They predicted high concentrations in the Arctic seas compared to concentrations in the subtropics, suggesting an increase in transport northwards which they attribute to Stokes drift in particular. Because Stokes drift has not been used universally in studies investigating microplastic transport, the study suggests microplastic contamination in Arctic regions could be more severe than currently expected. Winds and currents play important roles in the advection of plastic particles both into and out of the Arctic (Cózar et al., 2017), with macroplastic debris floating on the sea surface being affected by direct wind drag, which changes the drift dramatically compared to material that is fully submerged (Röhrs and Christensen, 2015). Obbard et al (2014) suggest that a large amount plastic pollution is entering the Arctic via the Pacific, where large concentrations of microplastics have been reported, and is subsequently being entrained within Arctic sea ice.

1.3.2 River transport. There have been very few studies thus far that have investigated Arctic rivers as a significant transport mechanism for plastic litter. Although there are numerous rivers that feed into Arctic coastal seas such as the Mackenzie and Yukon rivers in Canada and Northern America, and the Lena, Yenisey and Ob in Siberia (Holmes et al., 2013), inputs from these rivers are unlikely to contribute large amounts of plastic pollution to the Arctic as they

flow through sparsely populated areas (Obbard et al., 2014). However, Yakushev et al (2021) explain that the Arctic Ocean receives 11% of the global freshwater discharge, with the Ob (510 km<sup>3</sup>/year), Yenisei (630 km<sup>3</sup>/year) and Lena (530 km<sup>3</sup>/year) rivers providing the most input, suggesting that even with low concentrations, these rivers could be a significant source of plastic debris. In a study on the effect of Atlantic waters and Siberian rivers on microplastic distribution in the Eurasian Arctic, Yakushev et al (2021) found that smaller microplastics (with surface area <3 mm<sup>2</sup>) dominated in the low-saline inner plumes adjacent to the river estuaries and deltas, whilst in the outer plume microplastics had distinctly larger sizes (with surface area up to 10 mm<sup>2</sup>). Small differences were found between the average abundance of microplastics in the inner and outer plumes (4.5  $\times$  10<sup>-6</sup> and 5.1  $\times$  10<sup>-6</sup> particles L<sup>-1</sup> respectively), however when looking at the mass concentration, a notable difference was detected (1.6 μg m<sup>-3</sup> and 2.9 μg m<sup>-3</sup> respectively). The largest diversity in polymer types was found in the outer plume, consisting of five polymer types (13% fibres) compared to the inner plume which consisted of three polymer types (67% fibres). No microplastics were found outside of the river plumes in the saline Polar surface water mass. Yakushev et al (2021) explain that due to the absence of floating microplastics in this area, along with a higher abundance of subsurface microplastic fragments, it is presumed that released microplastics do not remain at the sea surface but sink to the subsurface layer after ice melting. This can be caused by physical and/or biological processes that modify floating properties of icetrapped microplastics and explains the absence of subsurface fragments of microplastics in river plumes. It is thought that floating microplastics in the plumes were discharged from the rivers to sea during an ice-free period and were not trapped by sea ice.

The study suggests the lower abundance of microplastics reported in the results compared with other investigations is possibly due to a difference in sampling procedures, and only considering particles >100  $\mu$ m to be comparable to previous research. The study also found the FT-IR analysis decreased the final number of particles identified as plastics, which is something all investigations into plastic pollution must consider. Yakushev et al (2021) go on to explain that the results found in this study are similar to levels reported for the Arctic Central Basin in Kanhai et al (2018), where similar sampling and identification methods were used, reinforcing the importance of consistent methodological approaches.

1.3.3 Sea ice transport. More recently, sea ice has been acknowledged as a significant sink for microplastic pollution, as well as a transport mechanism for moving plastic particles across the Arctic Ocean (Peeken et al., 2018, Mountford and Maqueda, 2021. Obbard et al., (2014) suggest that most of the plastic pollution entering the Arctic in surface currents is via the Pacific, where large concentrations of microplastics have been reported, and is subsequently being entrained to some extent within Arctic sea ice. Peeken et al (2018) suggest that sea ice is one of the key compartments for transporting and redistributing microplastics in the Arctic, but this will be dependent on prevailing ice floe drift patterns. In general, pack ice passes through different regions of the Central Arctic Ocean but is eventually carried southward to the Fram Strait via the Transpolar Drift. In order to determine drift trajectories and source areas of sampled sea ice, Peeken et al (2018) tracked sampled sea ice backwards in time using low-resolution ice drift via satellite observation, which showed that the sea ice originated from different source areas, namely the Amerasian and Eurasian Basins. The estimated time between plastic contamination and ice arrival varies throughout the Arctic. For example, contamination from sources in the Laptev and Kara seas take between 2-4 years to arrive in the Fram Strait, and up to 6-11 years from sources in the Amerasian Basin.

Mountford and Maqueda (2021) used a numerical model to look at the accumulation and transport of microplastics in the Arctic. They found that positively buoyant microplastics, arriving through surface transport, dominated Arctic sea ice. Whereas in comparison, they found that neutrally buoyant plastics transported to the area through deep water dominated sea ice in the Southern Ocean.

1.3.4 Bio-transport. The role of bio-transport poses two distinct issues: the first being marine biota acting as vectors for plastic and microplastic pollution. For example, ice algae has been suggested as a temporary sink and transport vector for microplastics (Bergmann et al., 2023). In a pilot study conducted in the Fram Strait, the ice algae *Melosira arctica* and sea water were sampled in three locations. Microplastics ( $\geq$ 2.2 µm) were detected in all samples, at concentrations ranging from 1.3 to  $5.7 \times 10^4$  microplastics m<sup>-3</sup> in ice algae and from 1.4 to 4.5  $\times$  10<sup>3</sup> microplastics m<sup>-3</sup> in sea water and comprised of 16 polymer types, with most particles (94%) being 10µm or smaller. During the melting of sea ice, ice algae detach from the sea ice and will either float in aggregates or sink rapidly to the ocean floor. The high concentrations of microplastics seen in ice algae in this study suggest it could act as an important transport

mechanism to the deep ocean floor where they become available to surrounding marine biota.

The second issue associated with bio-transport involves floating plastic debris acting as vectors for non-native species or pathogens that can enter the Arctic from other regions. For example, in Svalbard, non-native barnacles have been found on plastic debris (PAME, 2019). Macroplastic debris can act as 'rafts' transporting toxins and invasive species, posing risks to ecosystems across the Arctic, from both regional (i.e., within Arctic) and long-range sources (Barnes, 2002). Provencher et al (2010) studied ingested plastic in thick-billed murre (Uria lomvia), a migratory species to the Canadian Arctic, and reported the highest incidence of plastic ingestion to date for this species. Of the 186 thick-billed murres collected, 11% contained ingested plastics in their digestive tracts with a mean of 0.2 ± 0.8 microplastics found ingested per bird. The results of the study suggest that seasonal timing may determine how much plastic is ingested. For example, 13% of birds had ingested plastic early in the summer season when the birds had just arrived at the breeding colony. however, for birds sampled later in the season, after foraging in the remote area for several months, had no detectable plastics in their digestive tracts. Thick-billed murres may be ingesting plastics in the waters of the Northwest Atlantic during the winter months, where plastic contamination is relatively higher. During seasonal migration the birds will bring their burden of plastic contamination northwards to the Arctic. Presently, the quantities of plastic transported by bio-vectors such as migrating birds, fish and sea mammals into the Arctic is uncertain.

1.3.5 Atmospheric transport. Deposition with snowfall has been reported as a transport mechanism for smaller size ranges of microplastic particles. Bergmann et al (2019) used FT-IR to quantify microplastic pollution in 21 snow samples taken from ice floes in the Fram Strait, and 20 contained microplastic particles. The concentration of microplastics in these samples ranged from 0 to  $14.4 \times 10^3$  particles  $L^{-1}$  which is a substantial number but lower than concentrations reported in European snow where concentrations ranged from  $0.19 \times 10^3$  to  $154 \times 10^3$  particles  $L^{-1}$ . A variety of polymer types were observed in the snow samples, with acrylates/polyurethane-based varnish, rubber, polyethylene, and polyamide particles making up most of the concentration. Eighty percent of all detected microplastics were  $\le 25 \, \mu m$ , and 98% of all particles were  $< 100 \, \mu m$ . The detection limit in this study was  $11 \, \mu m$ , indicating large numbers of smaller particles could be missed, however techniques to identify particles this

small are lacking. The composition of the particles varied considerably, with 19 different polymer types found in total. The number of polymers per sample was significantly higher in European (mean,  $8.63\pm0.80$ ) compared with Arctic (mean,  $5.14\pm0.79$ ) samples, with acrylates/polyurethane-based varnish occurring most frequently (17 samples), followed by nitrile rubber, polyethylene, and polyamide. There was a high contrast in the polymer composition from European and Arctic samples, with much higher abundances of polyamide, acrylates/polyurethane-based varnish, rubber type 3, nitrile rubber, ethylene-vinyl-acetate, and PE in European samples. By contrast, polystyrene, polyvinyl chloride (PVC), polycarbonate, polylactic acid, and polyimide occurred exclusively in Arctic snow.

#### 1.4 Fate

Understanding contaminant fate within the Arctic Ocean is extremely difficult. This dynamic ecosystem is heavily influenced by the changing climate, which inevitably effects the transport and fate of contaminants such as plastic. At present, there have been few studies on the accumulation of plastics in the Arctic, especially when regarding sea ice and snow, which are vital features of the Arctic environment. Understanding the sources, transport, and fate of plastic pollution in the Arctic Ocean is critical to determining the potential threats posed by such contaminants to marine organisms that inhabit this ecosystem (Kanhai et al., 2020).

Reference	Location	Material	Sampling method	No. of samples	Size range (mm)	Lab methods	Microplastic concentrations		ntrations	Reporting unit
							Min	Max	Mean	
	Chuckchi Shelf	Ice	Sea ice core	1	0.22-5	Band saw, visual sorting and FTIR	n/a	n/a	~50	particles L <sup>-1</sup>
Obbard et al.	Siberian Sea	Ice	Sea ice core	1	0.22-5	Band saw, visual sorting and FTIR	n/a	n/a	234	particles L <sup>-1</sup>
(2014)	Beaufort Gyre	Ice	Sea ice core	1	0.22-5	Band saw, visual sorting and FTIR	n/a	n/a	38	particles L <sup>-1</sup>
	Laptev Sea	Ice	Sea ice core	1	0.22-5	Band saw, visual sorting and FTIR	n/a	n/a	~125	particles L <sup>-1</sup>
Peeken et al. (2018)	Fram Strait, north of Svalbard, Nansen Basin	Sea ice	Ice coring	5	0.011 - 0.15	Vacuum filtration and µFTIR (FPA detector)	(1.1 ± 0.8) x 10^6	(1.2 ± 1.4) x 10^7	n/a	Particles m <sup>-3</sup>
Bergmann et al. (2019)	Svalbard and Fram Strait	Snow	Surface sample with a spoon	14	0.011 - 0.475	Vacuum filtration (1 mL aliquot) and µFTIR	ND	14,000	1151.7 ± 3816.4	particles L <sup>-1</sup>
Geilfus et al. (2019) field study	Baltic Sea	Sea ice	Sea ice core	9	0.063 - 1.4	Vacuum filtration and Nile Red	8	41	n/a	particles L <sup>-1</sup>
Kanhai et al. (2020)	Arctic Central Basin (ACB)	Sea ice	Sea ice core	25	0.1 - 4.99	Vacuum filtration, visual sorting, and FTIR	2	17	6.46 ± 3.41	particles L <sup>-1</sup>

**Table 1.** Table showing studies on microplastic accumulation in Arctic and subarctic snow and ice; modified from Tirelli et al (2020).

1.4.1 Sea ice. The presence of plastic debris and microplastics in sea ice and the ice-rafted snowpack has been investigated in few studies to date. Bergmann et al (2016) sampled ice from both land-fast and drifting ice floes, to differentiate between local entrainment sources and long-range transport. Mean concentrations of  $2 \times 10^3$  particles L<sup>-1</sup> in pack ice and  $6 \times 10^2$ particles L<sup>-1</sup> in land-locked ice were detected. Eleven different polymer types were identified, with polyethylene being the most abundant. Calculation of drift trajectories by back-tracking of the ice floes sampled indicates multiple source areas, which explains the differences in the microplastic composition. Bergmann et al (2016) explain that the results from this study exceed concentrations from the North Pacific by several orders of magnitude, which can be explained partly by the process of ice formation, during which (organic) particles tend to concentrate by 1-2 orders of magnitude compared with ambient seawater. However, the magnitude of the difference suggests that Arctic sea ice is a temporal sink for microplastics. Increasing quantities of small plastic litter items on the seafloor nearby, corroborate the notion that melting sea ice releases entrained plastic particles, and that sea ice acts as a vector of transport both horizontally and vertically. Kanhai et al (2020) looked at microplastic concentrations in ice cores and seawater beneath ice floes and found that microplastic abundance in surface waters underlying ice floes (ND – 0.018 particles L<sup>-1</sup>) were orders of magnitude lower than microplastic concentrations in sea ice cores  $2 \times 10^{-6}$  to  $1.7 \times 10^{-5}$  particles L<sup>-1</sup>. The majority of the microplastics that were found in the ice cores were fibres (79%) and there was no consistent pattern between the vertical distribution of microplastics within the various sea ice cores. The origins of the ice were estimated by using backward drift trajectories, which showed that the ice possibly originated from the Siberian shelves, western and central Arctic.

To investigate the distribution and impacts of microplastics in sea ice, Geilfus et al (2019) conducted microcosm and field-based studies. Both approaches confirmed that sea ice does concentrate microplastics within its structure, with particles retained within the ice matrix. The microcosm study revealed that the presence of microplastics did not affect the growth and thickness of the sea ice, however, higher concentrations of microplastics were found where ice salinity was at its highest suggesting that brine mobility within the ice may affect the distribution of plastic particles within the ice column. The concentration of microplastics observed in the field study (8 – 41 particles L<sup>-1</sup>), which took place in the Baltic Sea, were one order of magnitude less than those used in the microcosm study. Although the concentrations observed by Geilfus et al (2019) were relatively low, there are concerns that higher concentrations of microplastics present in sea ice may alter surface sea ice properties such as the albedo. A reduction in ice/snow albedo due to the presence of particulate material like microplastics may promote surface melt.

Peeken et al (2018) sampled ice from the Fram Strait and Central Arctic to quantify the concentration and composition of microplastics. Their study found extremely high microplastic concentrations, with the highest particle concentration 1.2 ( $\pm$  1.4) × 10<sup>4</sup> particles L<sup>-1</sup> being observed in an ice core taken in the older pack ice of Fram Strait. These high concentrations are likely to reflect the age of the ice and the large geographic region through which the ice has drifted during its life cycle. The results from this study showed highly variable concentrations of microplastics in sea ice cores, with the second highest microplastics concentration found in the Greenland land-fast ice of the Fram Strait (core A; 4.1 ( $\pm$  2.0) × 10<sup>3</sup> particles L<sup>-1</sup>). Concentrations in cores collected north of Svalbard and in the Nansen Basin respectively, varied between 2.9 ( $\pm$  2.4) × 10<sup>3</sup> particles L<sup>-1</sup> and 2.4 ( $\pm$  1.0) × 10<sup>3</sup> particles L<sup>-1</sup>. The lowest concentration was found in a core taken from the north of Svalbard, 1.1 ( $\pm$  0.8) × 10<sup>3</sup> particles L<sup>-1</sup>. The values recorded in this study were 2-3 orders of magnitude higher than

in a previous study from the Central Arctic, where microplastic concentrations in sea ice were 0.038-0.234 particles  $L^{-1}$  (Obbard et al., 2014). The difference in results can largely be explained by the different methodology used. In the study by Obbard et al (2014), filters were first inspected using light microscopy and suspected microplastic particles were then analysed individually using FT-IR microscopy. In the Peeken et al (2018) study, Imaging FT-IR was used to scan entire areas of the filters, excluding the human bias which inevitably comes with visual selection of particles. Imaging FT-IR is able to detect very small particles (down to 11  $\mu$ m) which are most likely missed during visual inspection and are subsequently unreported. Peeken et al (2018) were able to show that the majority of microplastic particles identified in the sea ice cores were smaller than 50  $\mu$ m, and on average 67% of the particles were within the currently smallest detectable size class of 11  $\mu$ m, which would not have been considered in the Obbard et al (2014) study.

It is important to note that when comparing microplastic abundances in various studies, the methods used for sampling, quantifying, and identifying must be taken into consideration, as these can result in very different outcomes. Several studies have reported higher concentrations of microplastics in the smaller size ranges (e.g., Bergmann et al., 2017, Bergmann et al., 2019, Peeken et al., 2018), however certain sampling techniques may exclude these smaller size ranges, especially when looking at microplastics <100 μm, which may be leading to an underestimation of the abundance of microplastics in this region. Peeken et al (2018) found reported the highest concentrations thus far in Arctic sea ice, and the majority of microplastics identified using FTIR were <50  $\mu m$ , however, fibres were excluded from the analysis. In comparison, Kanhai et al (2020) reports a pre-dominance of fibrous microplastics (79%) in sea ice from the Arctic Ocean but did not include particles <100 µm due to limitations from the chosen analytical techniques (visual identification followed by FT-IR spectroscopy of potential microplastics). It is therefore apparent that if either fibres or particles <100 µm are excluded from results, microplastic concentrations in sea ice will be underestimated (Kanhai et al., 2020). As well as the methods used for identifying and quantifying, the units used to report plastic concentrations vary significantly, which adds to the difficulty of comparing results. As well as this, many studies do not report the procedures used to mitigate contamination in samples, through field sampling and lab processing, highlighting a pressing need for standardised reporting of quality control in plastic and

microplastic samples (Hallanger and Gabrielsen, 2018). It is vital that we create a more standardised regime that is widely accessible and generate clear data that can be shared amongst researchers to ensure a global picture of plastic pollution in our oceans and beyond is presented.

1.4.2 Surface and sub-surface waters. Oceanographic models have been used to show that once in the ocean, plastic debris can accumulate in large gyres (Cole et al., 2011). The fate of plastic particles there is not fully understood, but it is hypothesised that once in the gyres, plastic particles will be exposed to several processes such as biofouling, which may cause them to fragment until they are small enough to be ingested by marine biota. Cózar et al (2017) points out that the subtropical ocean gyres are recognised as accumulation zones for plastic debris, however because of their remote nature, the polar regions have been overlooked. In the study, the Arctic Ocean was highlighted as a 'dead end' for floating plastic debris. During a circumpolar expedition the Arctic Ocean was extensively sampled for floating plastic debris, and although little debris was found in Arctic waters, high concentrations of 6.5 g km<sup>2</sup> were found in areas of Greenland and the Barents Sea. The study explains that this sector of the Arctic Ocean plays a key role in the global Thermohaline Circulation (THC) through the formation of deep water by cooling, and as the THC actively advects warm surface water from low to high latitudes across the North Atlantic Ocean to the Arctic, it could collect buoyant plastic from highly populated latitudes, leading to accumulation in the Greenland and Barents seas. Here, the landmasses and polar ice cap create a 'dead end' for the surface transport of floating debris.

However, plastics have not only been found in surface seawater. Lusher et al (2015) found microplastics in both the surface (top 16 cm) and sub-surface (6 m depth) samples taken from Arctic polar waters. Microplastics were found in more than 90% of samples, both in the surface waters using a manta net and at 6m depth using the vessel's underway seawater pump. Surface sample concentrations ranged from ND -  $1.31 \times 10^{-3}$  particles L<sup>-1</sup> and averaged  $3.4 \times 10^{-4}$  ( $\pm 3.1 \times 10^{-4}$ ) particles L<sup>-1</sup>. Microplastic abundance in sub-surface samples ranged between ND -  $1.15 \times 10^{-2}$  particles L<sup>-1</sup> and averaged  $2.68 \times 10^{-3}$  ( $\pm 2.95 \times 10^{-3}$ ) particles L<sup>-1</sup>. Interestingly, the study found that although there were clear changes in polar fronts throughout the survey, microplastic abundances did not change with salinity, however there was a noticeable, although not statistically significant, difference with fewer particles in

colder, less saline waters. Particle composition comprised of 95% fibres, which they suggest were coming from the breakdown of larger plastic items used for shipping activity or fishing equipment, recreation, and offshore industries, and have likely arrived in the Arctic after long periods at sea, possibly transported over large distances. These microplastics may also be a result of the inputs from sewage and wastewater from more local sources e.g., washing of textiles. There was a high proportion of fibrous microplastics (> 90%) found in samples from Kanhai et al (2018) study on the abundance, distribution, and composition of microplastics in sub-surface waters of the Arctic Central Basin (ACB). Two independent sampling methods were used; underway sampling using the bow water system of ice-breaker Oden (single depth: 8.5 m), and conductivity, temperature, depth (CTD) profiler using a rosette sampler (multiple depths: 8-4369 m). Shallow, surface water samples contained microplastic accumulations ranging from non-detect to 7.5 x 10<sup>-3</sup> particles L<sup>-1</sup>, with CTD samples ranging from ND – 0.375 particles L<sup>-1</sup>. The results show that microplastics are present throughout the water column in the ACB, and that in natural conditions microplastics are being vertically transported out of surface waters. The highest levels of microplastics were reported near to the periphery of the ACB, in waters north of Svalbard. The study suggests this could be due to the lack of permanent sea ice cover in this region, allowing incoming Atlantic water to have a greater influence on near surface waters.

1.4.3 Deep-sea sediments. Deep sea sediments may be a permanent sink for microplastic particles that have deposited from surface waters following long-range transport via surface ocean currents or deposition from the atmosphere (Cózar et al., 2017, Kanhai et al., 2020). Bergmann et al (2017) found high quantities of microplastics in Arctic deep-sea sediments taken from the Long-Term Ecological Research (LTER) HAUSGARTEN observatory in the Fram Strait at 79°N, west of Svalbard (Norway), which currently comprises 21 sampling stations along a latitudinal and a bathymetric gradient between 250–5500 m water depth. Sediment samples were taken between 2340–5570 m depth, where an overall mean number of 4,356 particles kg<sup>-1</sup> sediment was found. The northernmost stations of this study, located within or close to the marginal ice zone, were found to have the highest microplastic abundances. A total of 18 different polymers were identified from all sediment samples, with almost 80% of particles being ≤20 µm diameter in size. The study found no significant correlation between depth and microplastic abundance, but a positive correlation was found between microplastic

abundance and chlorophyll *a* content, suggesting vertical export via incorporation in sinking algal aggregates. The study suggests that the overlying sea ice affects the quantity and release of microplastics present in the water column and hence their deposition to deeper waters and presence in bottom sediments. The study also corroborates previously discussed conclusions that Arctic sea ice may act as a transport vehicle for microplastics, as after the ice breaks up in the Central Arctic in spring it is transported to the south as ice floes with the Transpolar Drift. On route, it encounters warmer Atlantic surface waters and continues to melt, possibly releasing entrained microplastics.

Kanhai et al (2019) investigated deep-sea sediments of the Arctic Central Basin (ACB) as a potential sink for microplastics. Gravity and piston corers were used to retrieve sediments from depths of between 855-4353 m at 11 sites in the ACB. Surficial sediments from the various cores were taken and potential microplastics were isolated and analysed by Fourier Transform Infrared (FT-IR) spectroscopy. Of the surficial samples, 7 of the 11 samples contained synthetic polymers which included polyester, polystyrene, polyacrylonitrile, polypropylene, polyvinyl chloride, and polyamide. The study suggests that the presence of low-density polymers such as polypropylene and polystyrene indicate that there are mechanisms operating within the ACB that are affecting the density of microplastics, driving the vertical transport of these particles through the water column. Once plastics reach deepsea sediments, they are unlikely to become resuspended and redistributed, hence these areas being reported as significant sinks for microplastic pollution. There will likely be interactions between sinking particles and marine organisms inhabiting these areas, such as ingestion by benthic organisms. These organisms are highly sensitive to environmental change, relying on consistent and specific environmental conditions to survive, and so the addition of high concentrations of microplastics will inevitably have a detrimental effect on their health (Horton and Barnes, 2020).

#### 1.5 Summary

It is clear that the Arctic can no longer be considered as a pristine environment. Plastic debris including microplastics are present in the remote marine environment of the Arctic, although detrimental effects on marine organisms and wildlife, particularly through exposure to microplastics are not understood. The abundance of plastic pollution travelling to the Arctic

from long-range sources is difficult to measure against more localised sources, but the connectivity of Arctic marine areas with the rest of the world's oceans undoubtedly provides input from a range of distant sources (PAME, 2019). Macroplastics and microplastics arising through long-range sources can be transported via pathways such as ocean and atmospheric currents and can be transported within the Arctic circle through sea ice migration and biotransport. The fate of microplastics in the Arctic marine environment is still unclear, however their presence in seawater, sea ice, sediments and snow is unequivocal.

Unlike temperate regions, the cryosphere provides an additional environmental compartment that affects plastic distribution and fate in the Arctic. As highlighted in this chapter, sea ice has been recognised as a significant sink and potential transport vector for microplastic pollution, suggesting that high concentrations of microplastics are being transported across the Arctic and deposited in areas where we would not expect to find high abundances of plastic particles. Higher concentrations of microplastics in deep-sea sediments in regions with high ice cover (Bergmann et al., 2017) supports the view that ice is a significant source for microplastic pollution and may suggest these high concentrations in ice are exacerbating transfer to deep waters. However, at present, there is disparity on the number of plastic particles present in ice or seawater depending on the study in question and the analytical method involved. The reporting of ultra-small particles (e.g., <10 2m in diameter) can result in much greater particle number concentrations compared to studies that do not, and hence an increase in the number of observational studies across the Arctic for different environmental media is called for.

Ice provides habitats for sympagic (ice-associated) organisms, and it is currently unclear whether these organisms are being exposed to high concentrations of microplastics present in the ice. If current trends in sea ice melt continue, then over the next decade ~2.04 trillion m³ of ice will melt (Comiso, 2012). Taking the lowest observed particle concentration from Obbard et al (2014) (i.e., 38 particles per m³), would result in the remobilisation of > 1 trillion microplastic particles in Arctic Ocean surface waters. The entrapment and subsequent seasonal release of plastic debris and microplastic particles from sea ice has not been studied in any detail and the impact of this 'cycling' of plastic contamination on sympagic and pelagic organisms is unclear. One of the impacts from warming Arctic summers and the subsequent

reduction in sea ice cover, may be an increase in plastics debris in the Arctic, leading to an increase in plastic ingestion by marine biota (Mallory, 2008).

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# 2. The Beaufort Sea: an accumulation zone for microplastic pollution

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### **Abstract**

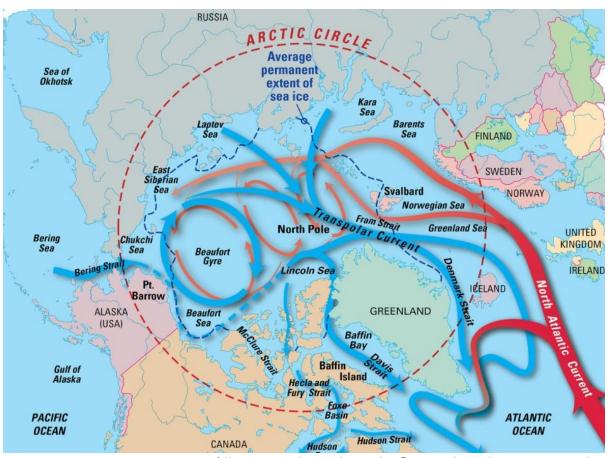
In the Canadian Arctic, the Beaufort Gyre has the potential to be a major sink for microplastic pollution, with Pacific and Atlantic currents bringing water and pollutants from long-range sources, as well as more localised sources including sea ice meltwater and riverine inputs. Surface (3 m) and sub-surface (100 m) water samples were taken from the Canadian Beaufort Sea in 2018, and microplastics were identified using µFT-IR analysis. Microplastic particles were found in all samples with concentrations ranging from 0.2 – 33.3 L<sup>-1</sup>. Interestingly, subsurface water samples (100 m depth) at several locations contained higher amounts of microplastics compared to surface water samples, indicating the influence of different water masses, although overall there was no significant difference in the average microplastic number concentrations between depths. The most common polymer types included polypropylene (88.8 %), polyethylene (5 %), and polyamide (2.6 %). These polymer types are consistent with those found in other areas of the Arctic, however, microplastic number concentrations in this study are much higher. This study highlights the need for further investigation of microplastics in the water column of the Beaufort Sea to improve our understanding of the sources, transfer, and fate of microplastic pollution in this region of the Arctic.

### 2.1 Introduction

The presence of plastic debris and microplastic particles in the Arctic marine system is of growing interest and concern (Bergmann et al., 2022). The Arctic is considered to be a significant sink for microplastic pollution, with notably high concentrations now observed in sea ice (Peeken et al., 2018). Importantly, plastic particles are present in deep-sea sediments of the central Arctic Ocean (Kanhai et al., 2019) with microplastic particles widely distributed

across the Arctic Ocean and throughout the water column (Tekman et al., 2020). The Beaufort Sea, located in the western Arctic, has the potential to be a significant sink for microplastic pollution in the Arctic. The Beaufort Gyre, centred in the Canada Basin of the Arctic Ocean, is a major reservoir of fresh water in the Arctic (Proshutinsky et al., 2019), where Pacific and Atlantic waters flow along the boundaries and are transported into the gyre through eddy processes. This creates several source pathways for microplastic pollution. Microplastics (≤ 5 mm in diameter) have been found in water samples across the Arctic (Lusher et al., 2015, Kanhai et al., 2020, D'Angelo et al., 2023), however there is significant spatial variation in particle concentrations across the Arctic as well as in the water column for different locations. To date, there have been very few studies looking at microplastics abundance in the Beaufort Sea and yet the Beaufort Sea displays seasonal variations in ice cover, temperature, light, freshwater, turbidity, and currents (Carmack and Macdonald, 2002). It provides habitat for a range of species, which have adapted to live through its environmental extremes. During winter months, the sea becomes covered by thick pack ice, an area of sea ice that is not attached to land, and in summer months, the sea ice melts northwards. This seasonal shift in ice cover triggers a surge in biological activity, leading to blooms of phytoplankton and zooplankton occurring initially along the boundaries, which form the foundation of the marine food web (Carmack and Macdonald, 2002).

There are several transport pathways leading to microplastic pollution in the Arctic, and specifically the Beaufort Sea, bringing plastics from both local and long-range sources (see Fig. 1). The surface waters of the Beaufort Sea are supplied by the Arctic and Pacific Oceans, with the deeper waters (> 200 m) originating in the Atlantic Ocean, increasing the likelihood of plastics entering the Arctic from sources further away. In the Canada Basin, the water column is split into three distinct layers. The first is the Polar Surface Water (0 - 250 m), comprised of the Polar Mixed Layer and the Pacific Halocline. The Polar Mixed Layer is the uppermost surface layer (0 - 50 m) and is characterised by low salinity water formed from the melting of sea ice and the influx of freshwater from riverine sources. Below this layer is a halocline (50 - 250 m), characterised by strong salinity which increases with depth and is made up of Pacific waters (Kanhai et al., 2018). Below this is the second layer, made up of Atlantic Water (250 - 900 m), and the third layer is Arctic Deep Water (900 m - seafloor) (Carmack and Macdonald., 2002, Kanhai et al., 2018).



**Figure 1.** Arctic currents map. (Illustration by Jack Cook, © Woods Hole Oceanographic Institution).

Another potential source of microplastics into the Beaufort Gyre is sea ice. Previous studies have highlighted sea ice as a significant repository of plastic particles and also a potential transport mechanism for microplastics in the Arctic (Peeken et al., 2018). Sea ice from the Beaufort, Chukchi, and East Siberian seas may become incorporated into the Beaufort gyre with some sea ice having the potential to exit this gyre and eventually join the Transpolar Drift, a major ocean current flowing into the Atlantic Ocean from the eastern Arctic (Kanhai et al., 2020, Peeken et al., 2018). In a study using numerical modelling, Mountford and Maqueda (2021) found that during the simulation, it was the Eurasian basin of the Arctic which showed the first signs of increasing levels of microplastics. They saw that over time, the buoyant microplastics trapped within sea ice were transported across the Barents Sea and north of Svalbard, as well as by the Transpolar Drift toward the Fram Strait. At the end of the 50-year simulation, the continued influx of microplastics via the Transpolar Drift resulted in the highest concentrations occurring north of Greenland, and towards Ellesmere Island and the Canadian Arctic Archipelago. An important local source of microplastics to this area could

be the concentration of shipping routes found at the margins of the Canadian Arctic Archipelago. As end-of-summer (September) sea ice declines (Serreze and Barrett, 2011), the frequency of ship visits is increasing (Adams et al., 2021).

Microplastics have also been reported throughout the water column in different regions of the Arctic (Kanhai et al., 2018, Lusher et al., 2015, Uurasjärvi et al., 2021). Deep sea sediments have been identified as a potential sink for microplastics in the marine environment (Kanhai et al., 2019, Zhang et al., 2020). There are several processes causing microplastics to sink, including physical and biological. Wind turbulence and storms can redistribute microplastics in the top layer of surface waters (Carmack and Macdonald, 2002), and biological processes such as feeding and excretion can result in microplastics being transported deeper into the water column, for example in the faecal pellets of zooplankton (Cole et al., 2016). Sinking ice algae aggregates have also been reported as a potential sink and transport pathway for microplastics to enter the deep ocean (Bergmann et al., 2023).

Few studies have looked at microplastic pollution in The Beaufort Sea. This study aims to quantify microplastics in surface and sub-surface water samples, to identify whether microplastic abundance varies at certain depths. Due to their low-density, we would expect to see more microplastic particles at the surface of the water. However, studies are increasingly reporting microplastic number concentrations throughout the water column. Sampling at these two depths should highlight whether these surface microplastics are migrating down to lower depths.

# 2.2 Materials & methods

2.2.1 Sample collection. Samples were collected during the Sikuliaq September 2018 cruise into the Beaufort Sea, North of Alaska, using a Rosette sampler. Samples were taken at 10 sites, from sub-surface (n = 10, average depth = 100.1 m) to surface (n = 10, average depth = 3.4 m), in Niskin bottles (12 L). Red cotton overalls were worn when sampling and filtering to prevent microplastic contamination. Samples were filtered from the Niskin bottle through 250  $\mu$ m plankton mesh, which were then encased in aluminium foil and placed in a -20 °C freezer.

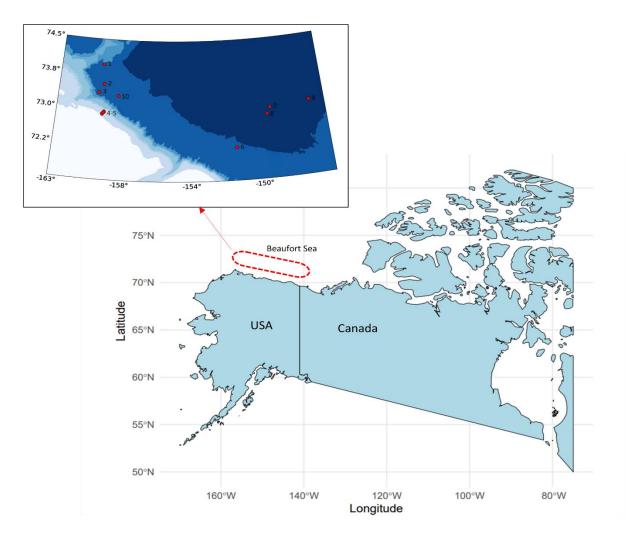


Figure 2. Map of the study area with focus map showing sampling area with stations labelled.

2.2.2 Sample processing and analysis. Samples were processed in a clean lab with monitored access. All work was undertaken in a laminar flow cabinet to mitigate any outside contamination, and cotton lab coats were worn at all times. Each mesh filter was attached to a metal frame using metal pegs and placed over a clean 1 L glass beaker. Deionised water and a natural fibre brush were used to rinse samples off the filters into the beakers. Samples in the glass beakers were then filtered through a clean steel mesh filter (27  $\mu$ m) to reduce the sample size. The samples were then rinsed off the steel mesh filter, back into the 1 L beaker, using Fenton's reagent (20 ml 30 % (0.3 kg<sup>-1</sup>) H<sub>2</sub>O<sub>2</sub> and 20 ml 0.05 M FeSO<sub>4</sub>), which was chosen due to it being an efficient method of removing organic matter whilst preserving the microplastics. These were checked regularly to monitor the reaction, and agitated every 1 hour, before being left for 24 hours. After 24 hours, samples were filtered onto clean steel mesh filters (27  $\mu$ m). These filters were then placed into 250 ml glass beakers and a shallow layer of 50 % (0.5 L<sup>-1</sup>) aqueous ethanol solution was added. Filters in ethanol were sonicated

for 10 minutes to release any particles from the filter. Using tweezers, the filters were lifted above the glass beakers, and using a glass pipette and ethanol the sample was rinsed off the filter. The samples were then poured into small glass vials, and a glass pipette was used to rinse beakers with ethanol to ensure all particles were transferred. Prior to analysis, each sample was carefully transferred onto 3  $\mu$ m, 25 mm silver membrane filters (Sterlitech<sup>TM</sup>) using a glass pipette.

Scanning  $\mu$ FT-IR (PerkinElmer Spotlight400, UK) analysis was performed in reflectance mode at a pixel resolution of 8 cm<sup>-1</sup> with 4 scans per pixel. An interferometer speed of 2.2 cm s<sup>-1</sup> was used, and scans were carried out from 4000 to 750 cm<sup>-1</sup> and included particles down to 25  $\mu$ m. Each sample took 89 minutes to complete. A microplastic identification software (siMPle v1.0.0) was then used to determine microplastic number concentration, as well as the polymer types and sizes of the microplastics found in the water samples. Each identified particle was assigned a correlation score, representing the similarity between the measured spectrum and the reference polymer. Only particles with correlation values  $\geq$  0.5 (or  $\geq$  50%) were accepted as confidently identified, and the majority of microplastics observed had a score between 0.6 and 0.8 (60 – 80%).

- 2.2.3 Statistical Analysis. Statistical analyses were performed using concentration data (particle  $L^{-1}$ ) in RStudio version 4.2.2 (Posit, 2022) using a significance level of p=0.05. Normality was tested using the Shapiro-Wilk test, before further statistical comparisons. Significant differences in microplastic number concentrations for depth were determined using the non-parametric Mann-Whitney U test as the data were not normally distributed.
- 2.2.4 Contamination mitigation and blanks. To minimise ship-based contamination, seawater sample filtration (from a 10 L non-plastic Niskin bottle on the ship's sampling rosette) was conducted on the open deck. Operators wore cotton-only overalls and gloves. In addition, aside from the nylon-filter mesh, all components of the sampling apparatus comprised of metal and glass. After water filtration the filter mesh was wrapped in aluminium foil and placed inside a zip-lock polyethylene bag and stored in a freezer (-20°C) prior to mesh extraction and processing. All reagents were filtered through GF/F filters (0.7 μm pore size) before use. All work was carried out in a laminar flow cabinet, and solvents and samples were covered with foil whenever they were removed (e.g., for weighing). 100 % cotton lab coats

were worn at all times. Blanks (n = 8) comprised of deionised water (1 L) passed through clean nylon mesh filters using the same procedures and processing steps as the samples.

There is currently no universal method of representing the values of microplastics found in blanks samples. For this study, microplastic counts were adjusted to compensate for the contamination in the blanks by subtracting the mean value of each polymer found in the blank samples from that polymer count in each of the environmental samples. Although there are limitations to this approach, applying a polymer specific average blank correction ensures that a consistent laboratory background contribution is removed from all environmental samples, reducing the risk of over-interpreting particles originating from contamination.

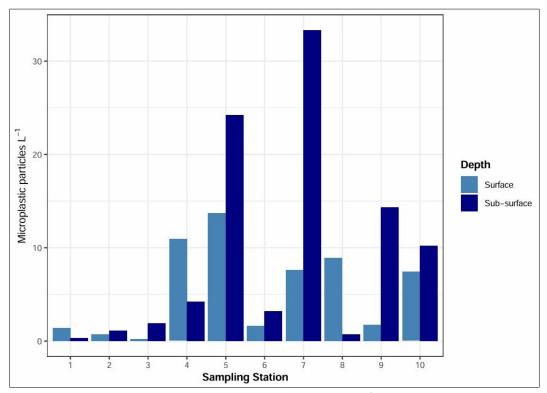
Despite these quality control measures, determining background contamination using procedural blanks remains challenging, particularly in Arctic environments where concentrations are often low. Procedural blanks cannot fully replicate all contamination pathways associated with ship-based sampling and laboratory processing, including airborne fibre deposition and handling during filtration. In the Beaufort Sea dataset, the occurrence of common polymers in blanks meant that background counts were sometimes comparable to those measured in environmental samples, increasing uncertainty in blank-corrected values. As a result, blank correction may either underrepresent laboratory contamination or disproportionately reduce low abundance environmental signals. These limitations require a cautious interpretation of corrected concentrations, which should be viewed as conservative estimates that reflect both environmental variability and unavoidable uncertainty in background contamination.

#### 2.3 Results

- 2.3.1 Quality controls. Blank data are provided in Table Supplementary 2. The average particle number for blanks was  $32.25 \pm 66.29 \, L^{-1}$ . 11 types of polymers were observed in blank samples, with polypropylene making up 77% of the total.
- 2.3.2 Microplastics in seawater. All water samples taken at 10 stations in the Beaufort Sea contained microplastics. Microplastics were observed in both surface (3 m) and sub-surface water (100 m) (Fig. 3), although there was no significant difference in the particle number concentration by depth (p = 0.597).

Surface samples had particle number concentrations that ranged between 0.2 and 13.7  $L^{-1}$ . Sub-surface samples showed higher particle number concentrations, ranging from 0.3 to 33.3  $L^{-1}$ .

Sampling stations 4 and 5 contained the highest particle number concentrations for surface samples; stations 5 and 7 contained the highest concentrations for sub-surface samples (Fig. 3).



**Figure 3.** Microplastic particle number concentrations (L<sup>-1</sup>) for surface and sub-surface samples at 10 sampling stations in the Beaufort Sea (see figure 2 for the approximate locations of the stations).

2.3.3 Microplastic polymer types. A wide range of polymer types were observed in both the surface and sub-surface water samples, with polypropylene making up vast majority with 88.8 % of particles comprising of this polymer (Fig. 4). Polyethylene made up 5 % and polyamide made up 2.6 %. Other polymer types included ethylene-vinyl-acetate (0.9 %), artificially modified cellulose (0.6 %), polyoxymethylene (0.6 %), acrylates (0.5 %), polyvinylchloride (0.4 %), polyester (0.2 %), polycaprolactone (0.1 %), polystyrene (0.1 %), polysulfone (0.1 %), and rubber (0.1 %).

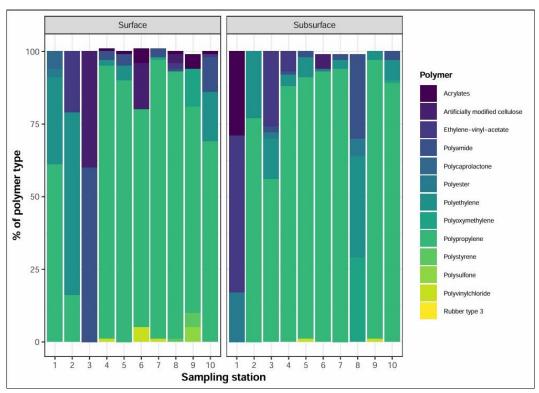
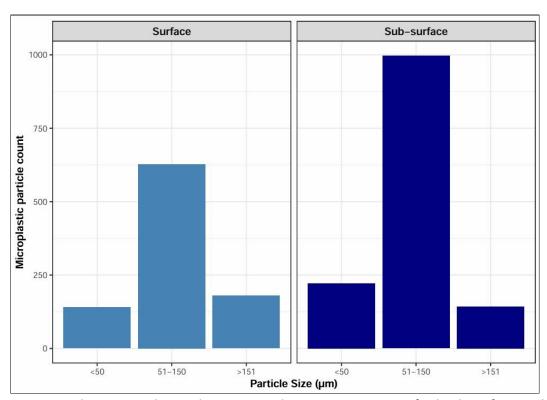


Figure 4. Fraction of polymer types found in each sample (%), split by surface and sub-surface.

2.3.4 Microplastic size. The majority of particles (70 %) were in the 51–150  $\mu$ m size range (Fig. 5). Although the plankton mesh used to filter the samples from the Niskin bottles was 250  $\mu$ m, we would expect to see smaller particles as it is likely they were entangled in organic matter on the filters or may have aggregated together during filtration. The smallest particles detected were 32  $\mu$ m, and all these particles were polypropylene. The largest particles found were > 10 mm in diameter and consisted entirely of polyethylene.



**Figure 5.** Microplastic particle numbers grouped into size categories for both surface and subsurface water samples.

2.3.5 Temperature and salinity. A clear difference was observed between temperature and salinity for the two sampling depths. The 3 m samples had an average temperature of  $-0.3\,^{\circ}\text{C}$  and salinity of 26.2, and the 100 m samples had an average temperature of  $-1.1\,^{\circ}\text{C}$  and salinity of 32.2.

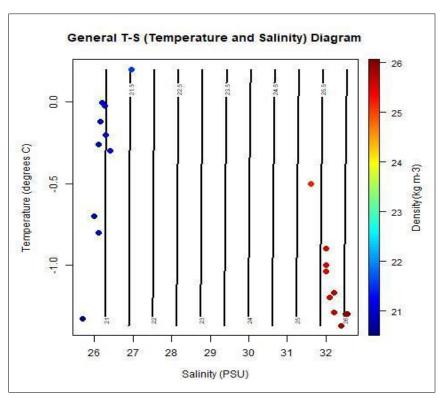


Figure 6. Temperature (°C) and salinity for surface (blue) and sub-surface (red) samples.

# 2.4 Discussion

2.4.1 Microplastics in the Beaufort Sea. Microplastic particle number concentrations in surface and sub-surface seawater samples from the Beaufort Sea are reported with a relatively wide range in concentrations (Fig. 3). To date, very few studies have looked at microplastic abundance in the Beaufort Sea in the Canada Basin, despite its location making it a likely sink for microplastic pollution from melting sea ice, riverine inputs, and Pacific waters.

In a study by Ross et al (2021), microplastic particles were characterised in 71 near-surface (3–8 m depth) seawater samples from across the Arctic. In the Western Arctic samples, the FT-IR confirmed microplastics averaged  $2.11 \times 10^{-2}$  ( $\pm 5.0 \times 10^{-3}$ ) particles L<sup>-1</sup>, of which 93.8% were fibres. Six sites in the Beaufort Sea were also sampled down to a depth of 1015 m. FT-IR confirmed microplastic counts from these samples averaged 3.73 x  $10^{-2}$  ( $\pm 6.9 \times 10^{-3}$ ) particles L<sup>-1</sup>, much lower than the concentrations found in this study. Another study found an average microplastic concentration of 1 ( $\pm$  1) L<sup>-1</sup> in the Beaufort Sea (D'Angelo et al., 2023). Microplastic concentrations have also been reported for other areas of the Arctic. In the Fram Strait, microplastic concentrations in seawater samples were 0.0014 – 4.5 particles L<sup>-1</sup>

(Bergmann et al., 2023). In the Barents Sea and Central Arctic, average microplastic abundance was reported as  $8.5 \times 10^{-4}$  and  $7.8 \times 10^{-4}$  items L<sup>-1</sup> respectively (Pakhomova et al., 2022). In a study by Barrows et al (2018), surface waters of the Arctic ocean contained the highest microplastic concentrations of  $31.3 \pm 6.5$  particles L<sup>-1</sup>. Results from this study show that microplastics were identified in all seawater samples, both surface (3 m) and sub-surface (100 m) using  $\mu$ FT-IR, at much higher concentrations than those reported in other studies.

The Beaufort Sea is a complex area of the Arctic where multiple transport pathways for pollution come together. The wind-driven Beaufort Gyre, a clockwise circulation system centred over the abyssal plain of the Canada Basin, dominates the mean, large-scale movement of sea ice and surface water (Fig. 1). However, below the surface waters, the flow changes to anticlockwise, forming the Beaufort undercurrent. This flow moves both Pacific and Atlantic waters eastward along the continental margin (Carmack and Macdonald, 2002). The circling motion of ocean currents and wind patterns has the potential to create a sink or 'hot spot' for plastic pollution, and many ocean gyres around the world have been reported to have high levels of microplastics contamination (Jiang et al., 2020, Lebreton et al., 2018, Poulain et al., 2018). In a study by Lima et al (2021) the Arctic is highlighted as a potential accumulation zone for microplastics using spatial modelling. The results predicted microplastics from the North Pacific Gyre and from the subpolar Pacific Gyre are being flushed towards the Arctic, and once there, the accumulation of microplastics occurs outside the Beaufort Gyre, since it is a zone of accumulation of sea ice and freshwater.

Ocean currents are not the only process potentially transporting microplastics to this area of the Arctic. The Beaufort Sea, specifically the Canadian Shelf, is subject to extreme annual and interannual variations in its river inflow (Carmack and Macdonald, 2002). Most of this river discharge (60 %) comes from the Mackenzie River, highlighting it as a potentially significant transport mechanism for microplastics originating in Canada (Adams et al., 2021). This would be at its most effective during the spring thaw, when the river flow is at its peak (Tirelli et al., 2020). Adams et al (2021) attributed the higher concentrations of microplastics found in samples from the Beaufort Sea to the nearby Mackenzie River, which can transport microplastics deposited atmospherically and sourced from within the drainage basin. The river encompasses a region with a population of around 400 000, which is heavily industrialised.

A rapidly warming Arctic is likely to alter both the sources and distribution pathways of microplastics in ways that mainly exacerbate the patterns observed in this study. Ongoing reductions in sea ice extent and thickness mean that one of the key temporary sinks for microplastics is diminishing, potentially shifting the system from storage to release, particularly during earlier and more prolonged melt seasons. This may enhance the delivery of microplastics to the surface and subsurface waters sampled here, increasing the subsurface accumulation identified in this chapter. At the same time, increased river discharge and expanding shipping and industrial activity are expected to introduce additional microplastic sources to the region. Changes in large-scale circulation, including a potential weakening or re-organisation of the Beaufort Gyre (Timmermans et al., 2025), could further modify the residence time and transport pathways of microplastics, potentially reinforcing the retention of particles within the water masses where higher concentrations were detected. While some mechanisms, such as enhanced flushing of the upper ocean with meltwater, could locally dilute particles, the net effect of Arctic amplification is projected to increase the availability, mobility, and persistence of microplastics.

2.4.2 Microplastics in the water column. The distribution of microplastics within the water column is changeable due to the influence of multiple factors, including wind, tides, and current circulation (Kanhai et al., 2018). The salinity of the sampled seawater (Fig. 6) indicates that the 3.4 m-samples were taken from Polar Surface Water (average temperature and salinity of -0.3 °C and S = 26.2) and the 100 m-samples from the Pacific Halocline (-1.1 °C, S = 32.2). Salinity, temperature and density variations indicate that different water masses were sampled and hence may reflect different sources of microplastics (Liu et al., 2024). Unlike most of the world's oceans, water density in the upper Arctic Ocean is primarily controlled by salinity (Mackinnon et al., 2021). Lusher et al (2015), found that less saline (fresher) nearshore sub-surface samples taken near Svalbard were free of microplastics, likely because unpolluted freshwater diluted microplastic concentrations, whereas more saline offshore waters contained Atlantic water polluted by long-distance transport of microplastics from more populated areas to the Arctic. These factors also determine the density of seawater, and we would expect to see microplastics settling to depths at which they are neutrally buoyant. However, there are a range of physical and biological factors that can cause this to change.

The difference between the mean particle concentrations for the surface versus deeper waters was not statistically significant. This may suggest that the particles have originated from the same source.

2.4.2.1 Microplastics in surface samples. Microplastics contained in surface samples from the Polar Mixed Layer likely originated from melting sea ice, or possibly freshwater influx from the Mackenzie River, which could transport microplastics to the Beaufort Sea from heavily industrialised areas. The peak flow of the Mackenzie River occurs earlier in the year during the spring thaw; however, it is still possible microplastics could enter the Polar Mixed Layer via river inflow but could explain why microplastic numbers are relatively low in surface samples. It may also be that microplastic numbers are being diluted by unpolluted freshwater rather than increased (Lusher et al., 2015).

Sea ice has been shown to harbour higher concentrations of microplastics compared to the surrounding seawater. This is most likely due to particle entrainment during ice growth and accumulation of particles deposited from the atmosphere over the winter with, for example, snowfall. Upon melting, sea ice becomes a significant source of microplastics. In Chapter 3, (Ball et al., unpublished) laboratory-based sea ice simulations show that microplastics accumulate at the uppermost layer of ice, and when the ice melts, these particles are abruptly released into the surrounding surface water. In the Arctic, the melt season occurs between June and September and during this time, fresher melt water is returned to the surface ocean (Timmermans and Toole, 2023). Samples in this study were taken during September, when the Polar Mixed Layer is shallower and less saline compared to winter and particles released from ice melt have become more widely distributed and/or descended further down the water column (Kanhai et al., 2020, Peeken et al., 2018). Carmack and Macdonald (2002) explained that below 20 m there often remains a cold, saline remnant of the previous year's polar mixed layer, which may retain microplastics, therefore acting as another source.

2.4.2.2 Microplastics in subsurface samples. High microplastic concentrations were observed in sub-surface samples at several sample locations. The highest sub-surface concentrations were observed at sampling stations 5 (shelf break;  $24.2 L^{-1}$ ) and 7 (northeast of ice sheet near Marginal Ice Zone (MIZ);  $33.3 L^{-1}$ ). Location may influence the amount of microplastics found in samples and be representative of different water masses. In a study by Von Friesen et al (2020) which looked at anthropogenic microparticles (including microplastics) in Svalbard, the

highest microplastic concentrations were also found in sub-surface water samples from the MIZ, which they explain are likely released during the melting of sea ice. At stations 4 and 5, located at the shelf break, surface samples contained higher concentrations. Here, the maximum depth was 144 m at station 4 and 181 m at station 5, meaning the sub-surface samples taken at these stations were almost at the seafloor. Water here is flowing off the continent into deeper waters, and is prone to shelf-break upwelling, where surface waters can become displaced by strong winds and are replaced by cold, nutrient rich water that comes up from below. Carmack and Macdonald (2002) explain that a portion of the freshwater can be forced off the shelf under certain wind conditions, and in turn this is then replaced by saltier water from the interior ocean. This saltier water may bring with it microplastics from other areas of the Arctic, adding to the microplastic burden at these sites.

There are several reasons as to why microplastics may be found in deeper waters. Studies have shown that microplastics can be ingested by marine organisms present in surface waters and be transported to deeper waters through biological processes such as migration, feeding, and excretion. Microplastics have been found in the faecal pellets of zooplankton (Cole et al., 2016, Gunaalan et al., 2023), and in the digestive tracts of Beluga Whales in the Eastern Beaufort Sea (Moore et al., 2020). Ice algae, which has been reported as a potential sink for microplastic pollution (Bergmann et al., 2023), account for 10-15 % of the primary production for the Canadian shelf (Carmack and Macdonald, 2002). If the algal mats are not sufficiently grazed, they can separate from the ice and subsequently sink. This provides almost all the carbon flux that nourishes the benthos (Carmack and Macdonald, 2002), but may also act as a transport mechanism, moving microplastic particles further into the water column with subsequent deposition to the seafloor. Physical processes such as turbulence and mixing in the water column may also lead to microplastics sinking deeper into the water column or stratification with certain water masses. In the Beaufort Sea, storms throughout summer and early autumn can mix the top 10 to 20 m of the water column (Carmack and Macdonald, 2002), which could lead to the redistribution of plastic particles.

Alternatively, the main source of microplastics found in sub-surface samples could be long-range transport with ocean currents. Water samples taken at a depth of 100 m, represent the Pacific Halocline layer of the Polar Surface Water. Here, water from the Pacific flows in through the Bering Strait to the Beaufort Sea and exits via the Transpolar Drift, through the

Fram Strait and Greenland Sea (Aksenov et al., 2016, Kanhai et al., 2018). It is probable that microplastics found in these samples have originated from the Pacific Ocean. Some of the highest microplastic levels have been observed in the North Pacific Ocean and its marginal seas (Shim and Thompson, 2015). The eastern part of the North Pacific Subtropical Gyre, also known as the 'Great Pacific Garbage Patch', is thought to contain at least 79 thousand tonnes of ocean plastic inside an area of 1.6 million km². The Bering Strait has been highlighted as a pathway for microplastics being flushed from the North Pacific Gyre and subpolar Pacific gyre towards the Arctic by the Alaska Coastal Current (Lima et al., 2021). Microplastics have also been found in the Bering and Chukchi seas, with higher concentrations found in the latter, ranging from 8.6 x 10-5 to 3.1 x 10-4 particles L-1 (Mu et al., 2019). D'Angelo et al (2023) highlighted the importance of the Barrow Canyon on the distribution of water masses. suggesting that water transport through this seabed acts as a major pathway for microplastic transport. Suspended microplastics maybe directed deeper into the Beaufort Basin where shelf break processes may cause microplastics to either sink to the seafloor or flow to the surface.

2.4.3 Microplastic polymer types and size. The most common polymer types found in the North Pacific Subtropical Gyre include polyethylene and polypropylene, likely originating from containers, bottles, rope, and fishing nets (Lebreton et al., 2018). In the Bering and Chukchi Seas, polymer composition consisted mostly of polyethylene terephthalate, polypropylene, and polyethylene (Mu et al., 2019). Sea ice and seawater samples from the Canadian Arctic Archipelago and Western Arctic marginal seas also showed an abundance of polyvinyl chloride (D'Angelo et al., 2023) with a relatively low diversity of polymer types.

The range of polymers identified in this study suggest multiple sources. However, the most common polymers were polypropylene (88.8 %), polyethylene (5 %), and polyamide (2.6 %) which are some of the most common polymers used in plastic production and are subsequently found in most samples from the Arctic (Bergmann et al., 2017, Lebreton et al., 2018, Peeken et al., 2018, Ross et al., 2021, Tekman et al., 2020). These polymers are used in a wide range of applications including polyamides, e.g. nylon, used in fishing lines and nets (BPF, 2024). Polyethylene is also one of the most widely used plastics in the world and is made into products ranging from food packaging and shopping bags to car fuel tanks (Encyclopaedia Britannica, 2024). Polypropylene is used in many plastic products which require toughness,

flexibility, light weight, and heat resistance. It can also be spun into fibres used in home furnishings and textiles (Encyclopaedia Britannica, 2024). It has a density between 0.895 and 0.92 g cm<sup>-3</sup>, making it the commodity plastic with the lowest density (<u>Plastics Europe</u>). The abundance of this low-density polymer in sub-surface samples strongly suggests these particles originated in the surface layer but have sunk over time, due to physical or biological processes. As the polymer composition did not differ with depth (i.e. dominated by polypropylene) then this suggests that the microplastics originated from similar sources.

The size of microplastics reported in this study were limited by the sampling method, whereby samples were filtered through a 250  $\mu$ m plankton mesh. This does not, however, exclude smaller size ranges as is it possible that smaller microplastics could have been trapped by organic matter on the mesh filters, but we must assume that there were potential losses of smaller microplastics and results may be an underrepresentation. The majority of microplastics (70 %) were found in the size category 51–150  $\mu$ m (Fig. 5). Several Arctic studies have reported higher concentrations in smaller size ranges (Bergmann et al., 2017, Bergmann et al., 2019, Peeken et al., 2018) which are likely attributed to different sampling and processing procedures relative to this study. Microplastics found in beluga whales from the Eastern Beaufort Sea were dominated by smaller size ranges (< 500  $\mu$ m) (Moore et al., 2020). However, there is little information on the quantity of microplastics in this area, let alone the characteristics of these particles. It is also important to note that comparing results of microplastic abundance in different studies is made difficult by the use of different sampling and processing techniques, which may exclude certain sizes and shapes of microplastic particles (Pakhomova et al., 2022).

# 2.5 Conclusion

This study demonstrated microplastic abundance in two distinct layers of the Beaufort Sea: the Polar Mixed Layer (3 m), and sub-surface (100 m) in the Pacific Halocline. The sampling area and depths provide an insight into the transport and fate of microplastics in the Beaufort Sea. There are a range of potential sources feeding into this water system, however results from this study strongly suggest that microplastics in both surface and sub-surface samples originated from freshwater inputs or sea ice melt in the surface layer, and that various physical and biological processes have caused them to sink. Polymer composition did not vary

between depths, further supporting the conclusion that microplastics in these samples originated from the same source. The highest microplastic numbers were found at the shelf break and near the MIZ, highlighting the importance of spatial variability in microplastics abundance. This study provides evidence of microplastic accumulation in the Beaufort Sea, and that there are multiple transport pathways driving this pollution. The abundance of microplastic particles in the 100-m samples suggests microplastics may be sinking to the seafloor. Future research should focus on the accumulation of microplastics in the deep-sea sediments of the Beaufort Sea.

# **Acknowledgements**

The samples used in this study were collected onboard the Sikuliaq research cruise during September 2018 in collaboration with the Changing Arctic Oceans PEANUTS project by Ben Barton. I would like to thank Yueng-Djern Lenn for providing essential data and knowledge used in this chapter. Laboratory analysis of these samples was conducted at Lancaster University. I would like to thank Nhu Phan for her help during the processing of these samples. Microplastic identification in this chapter was conducted at NOC Southampton. I would specifically like to thank Alice Horton and Katsia Pabortsava for their support during this work.

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# **Supporting Information**

Table S1: Depth (m), coordinates, volume (L), weight (g), total microplastic count, blank corrected microplastic counts, and microplastic number concentration ( $L^{-1}$ ) for all samples.

Station	Sample	Lat	Long	Volume	Sample	Total	Blank	MP
	depth			filtered	weight (g)	MP	corrected	number
	(m)			(L)		count		concentrati
								on (L <sup>-1</sup> )
1	3.3	73.961	-160.08	11.89	211.614	47	16.75	1.4
1	99.7	73.961	-160.08	11.59	154.254	28	3	0.3
2	3.5	73.538	-159.95	11.87	260.97	38	7.875	0.7
2	100.1	73.538	-159.95	11.82	246.13	42	13.25	1.1
3	4.4	73.364	-160.25	11.889	301.74	12	2.5	0.2
3	100.5	73.364	-160.25	11.799	275.18	53	21.875	1.9
4	3.5	72.931	-159.89	11.95	297.47	161	130.5	10.9
4	100.2	72.931	-159.89	11.99	284.06	80	50.375	4.2
5	3.4	72.959	-159.83	12	360.23	196	164.75	13.7
5	100.3	72.959	-159.83	11.145	408.05	299	269.5	24.2
6	3.0	72.206	-151.08	11.83	406.71	47	19	1.6
6	99.6	72.206	-151.08	11.9	480.93	65	37.75	3.2
7	3.3	73.017	-148.68	11.92	425.61	121	90.125	7.6
7	100.3	73.017	-148.68	11.89	478.82	427	395.75	33.3
8	3.3	72.878	-148.91	11.87	402.47	136	105.25	8.9
8	100.1	72.878	-148.91	11.83	340.07	38	8.5	0.7
9	3.1	73.083	-146.02	10.96	373.83	48	18.625	1.7
9	100.0	73.083	-146.02	11.865	461.22	200	169.75	14.3
10	3.4	73.312	-158.93	11.9	536.56	118	88.25	7.4
10	100.3	73.312	-158.93	11.89	344.43	151	120.75	10.2

Table S2: Microplastic concentrations and polymer types for blank samples.

Polymer type	Number of particles	Average (particle
	found in blanks (n = 8)	no. / 8) P L <sup>-1</sup>

acrylates/polyurethanes/varnish	1	0.125
cellulose artificial modified	8	1
polyamide	4	0.5
polyester	4	0.5
polyethylene	32	4
polyethylene oxidized	1	0.125
polypropylene	198	24.75
ethylene-vinyl-acetate	3	0.375
polyoxymethylene	4	0.5
polyvinylchloride	1	0.125
rubber type 3	2	0.25
Total	258	32.25

Table S3: Temperature, Salinity and Dissolved Oxygen information at each depth for each station.

Station	Depth	Temp (°C)	Salinity (PSU)	Dissolved Oxygen
				(umol / kg)
1	Surface	-0.3	26.4	363.988
1	Subsurface	-0.5	31.6	321.008
2	Surface	-0.26	26.1	364.767
2	Subsurface	-0.9	32	306.4
3	Surface	-0.2	26.3	363.206
3	Subsurface	-1.17	32.2	305.451
4	Surface	-0.02	26.26	367.255
4	Subsurface	-1.3	32.5	277.561
5	Surface	-0.12	26.14	369.53
5	Subsurface	-1.3	32.56	300.782
6	Surface	0.2	26.95	362.088
6	Subsurface	-1.37	32.4	292.001
7	Surface	-0.7	26	375.115
7	Subsurface	-1.2	32.1	303.501
8	Surface	-1.33	25.7	380.655

8	Subsurface	-1.29	32.2	300.235
9	Surface	-0.0027	26.2	366.27
9	Subsurface	-1.04	32	308.912
10	Surface	-0.8	26.1	375.207
10	Subsurface	-1	32	307.869

# 3. Mechanistic understanding of microplastic fate in sea ice: a simulated freeze – thaw study.

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#### **Abstract**

Microplastic particles are present in the surface waters of the Arctic Ocean and have been observed in sea ice across large regions of the Arctic. However, the role of sea ice in accumulating plastic particles, as well as serving as a temporary sink and vector of plastics is not well understood. In this study, sea ice was grown in a sea ice chamber from seawater that was artificially spiked with microplastic polyethylene spheres, to investigate plastic particle uptake into ice and their fate following ice growth and subsequent melt. Experiments were conducted whereby ice was grown to a depth of ~15cm before the onset of a thaw period to induce ice melt. During each freeze period, plastic particles in the seawater (Freeze 1 average: 1.28 x 10<sup>4</sup> particles L<sup>-1</sup>, Freeze 2 average: 9.75 x 10<sup>3</sup> particles L<sup>-1</sup>) became entrained into the ice, with the highest particle numbers in the uppermost ice layers (furthest from the seawater) and dominated by the smallest size particles (orange spheres, 125 - 150 μm). Enrichment of particles in the ice relative to the surface seawater was observed ( $EF_{MP} = 12$ ) indicating that low density particles (with neutral or positive buoyancy in seawater) are readily entrained into new and growing sea ice and appear to have a vertical distribution in the ice that is similar to the distribution of ice brine. Following the onset of ice melt, plastic particles were released from the ice and accumulated in a less-saline buoyant meltwater layer. This raises the possibility of increased exposure to plastic particles for sympagic organisms during seasonal ice melt in the Arctic.

# 3.1 Introduction

Due to their low-density and small size, microplastics are subject to long range environment transport in air and ocean currents and have now reached some of the most remote areas of

the world, including the Polar regions (Obbard, 2018), where local sources of marine pollution are minimal. Polar regions are increasingly receiving plastic waste (Horton and Barnes, 2020), and this is continuing to rise as populations grow and tourism increases (Polasek et al., 2017). As well as being transported across large regions, microplastics are able to move from the ocean surface to deep waters and marine sediments via gravitational settling or more complex routes e.g., sinking ice algae aggregates (Bergmann et al., 2023). However, this movement, as well as their fate and behaviour in remote marine environments, is not well understood.

Microplastics have been discovered in both surface and subsurface water samples in the Arctic, proving they are readily available to sea ice (Lusher et al., 2015), which has been identified as a potential seasonal sink for microplastic particles (Peeken et al., 2018, Mountford and Maqueda, 2021). However, the presence of plastic debris and microplastics in sea ice and the ice-rafted snowpack has been investigated in only a few studies to date (Ball and Halsall, 2023). Currently, there's a lack of knowledge concerning the processes and rates of microplastic incorporation within sea ice, and the possible effect on sea ice properties, such as influencing the albedo of surface snow (Geilfus et al., 2019).

In the Arctic, seawater freezes to form sea ice during the winter months. As the air and surface seawater temperatures drop, freshwater ice crystals separate from saltier seawater and accrete from the surface downward (Obbard et al., 2014). During the initial stages of ice formation, when small (< 1 mm) frazil ice crystals aggregate and grow, particulate matter in the surrounding seawater can become entrapped in the ice crystal structure. Persistent organic pollutants such as polychlorinated biphenyls (PCBs) have been shown to accumulate in sea ice (Garnett et al., 2019). During a laboratory-controlled experiment, sea ice formation was shown to result in the entrainment of chemicals dissolved in the seawater, and concentration profiles in bulk ice showed the highest levels in both the upper (ice–atmosphere interface) and lower (ice–ocean interface) ice layers, suggesting their incorporation and distribution is influenced by brine advection within the growing ice column. It was suggested that the movement of brine within sea ice could control the fate of these chemicals and provide a pathway for exposure to ice-associated biota. This is likely to be the case for fine particulate matter like microplastics.

Microplastics can become encapsulated in the sea ice as it freezes, forming a matrix of ice with embedded plastic particles. Once entrapped, microplastics can be advected as ice sheets move through circumpolar drift patterns. When the sea ice melts during warmer seasons, the microplastics will be released back into the water. In a study using numerical modelling, Mountford and Maqueda (2021) looked at sea ice as a possible repository for positively buoyant microplastics. They demonstrated a seasonal cycle of capture and release of plastics, with a consistent rise and fall in the quantities of microplastics trapped within the sea ice. After a rapid increase over approximately the first 15 years of the simulation, the ratio of the mass of plastics in sea ice appeared to reach a steady state, despite the mass of plastics in sea ice and surrounding water continuing to increase. It was suggested that this could indicate a "capacity" for the proportion of microplastics that can be sequestered from the underlying water, which must be dependent on the rates of sea ice formation and melt. Although microplastic incorporation at the surface of sea ice is not thought to affect sea ice growth, there are concerns for ice salinity and changes in ice albedo, which may promote surface melt (Geilfus et al., 2019).

The Arctic landscape is changing, with a warming climate resulting in a significant decline in the areal extent of sea ice cover (Meredith et al., 2019). This sea ice cover is also getting much younger, primarily consisting of first year ice (ice that is less than one year old) (Stroeve and Notz, 2018). If the current trend of melting sea ice continues, approximately  $2.04 \times 10^{12}$  m³ of ice is estimated to melt over the next decade (Comiso, 2012). Considering the lowest observed sea ice particle concentration of 0.038 L<sup>-1</sup> (Obbard et al., 2014), this would result in the release of over 1 trillion microplastic particles into the surface waters of the Arctic Ocean. Currently, there's a lack of knowledge concerning the occurrence, accumulation, and 'cycling' of microplastics with sea ice, and their possible effect on sea ice properties. Studies have previously focused on identifying the abundance, sources, and accumulation patterns of microplastics, but the effects that plastic pollution is having on Arctic ecosystems is unknown (Horton and Barnes, 2020).

This study aims to understand microplastic uptake into forming sea ice from seawater, to investigate whether accumulation in ice is apparent and to track particle fate during simulated seasonal thaw. Due to the low-density and small size of the particles, we hypothesise that the highest concentration of particles will be found in the uppermost layer of sea ice and that

enrichment of particles in sea ice (relevant to the underlying seawater) will be apparent. We anticipate that the findings from this study could be used to provide a mechanistic understanding of microplastic uptake behaviour within sea ice, and to better understand how sympagic organisms could be exposed to microplastics during periods of brine expulsion or ice melt.

# 3.2 Materials and Methods

3.2.1 Experimental setup. A simulated freeze up – thaw down experiment was conducted in the Roland von Glasow Air-Sea-Ice Chamber (RvG-ASIC) at the University of East Anglia, Norwich, UK. The facility consists of an insulated glass-walled tank (dimensions: height: 1.2 m; width 1.2 m; length 2.5 m) located inside a refrigerated chamber (the air can be chilled to -50°C). 'Artificial seawater', which will be referred to as seawater throughout this chapter, was made by dissolving NaCl in deionised water to a concentration of  $\approx$ 35 g L<sup>-1</sup> (all volume concentrations are reported for 20 °C). A submerged pump was used to continuously mix the seawater throughout each experiment when ice was not present, and a series of digital thermometers measured the in-situ sea ice temperature profile and calculated the sea ice depth. Two freeze–thaw experiments, referred to as "Freeze 1" and "Freeze 2" hereafter, were performed at air temperatures of (–25 ± 1) °C and (–30 ± 1) °C, respectively.

A range of low-density polyethylene spheres were used to simulate microplastic pollution in the sea ice chamber and are detailed in Table 1. Although a range of polymers would have been a better representation of microplastics found in the Arctic, polyethylene has been shown to be a dominant particle in many studies sampling Arctic waters and sea ice, and the polyethylene spheres used for this study were available in a range of sizes and fluorescent colours which were necessary for the design of this experiment. A range of sizes and densities were used in an attempt to show a more accurate representation.

The expected initial particle number concentrations before freeze-up, were calculated based on the mass of each particle type added to the water and with knowledge of the mean particle radius as well as assuming the particles were homogeneously distributed over the entire water volume of the chamber i.e. 3300 L. The resulting concentration (particles L<sup>-1</sup>) for each particle type was determined by spiking a known mass of each particle type into the chamber

seawater and assuming homogeneous mixing. The latter was achieved through 24-hour water mixing using water pumps.

The concentrations of microplastic particles introduced into the chamber were selected to ensure both reliable recovery during sampling and environmental relevance. Using very small spike quantities risked producing particle abundances below the detection limits of the sampling and analytical methods used in this study. I therefore opted for a concentration high enough to guarantee measurable particle recovery across all compartments of the system, while avoiding unrealistically elevated loads. Importantly, reported concentrations of microplastics in the Arctic vary widely between studies and sampling approaches, with some observations documenting exceptionally high concentrations in surface and subsurface waters and ice samples for the smaller size fractions most comparable to the particles used here. This approach therefore attempts to balance practical experimental requirements with consistency to reported findings in Arctic samples.

Colour	Mass (g)	Diameter range (μm)	Density (g cm <sup>-</sup> <sup>3</sup> )	Concentration (L <sup>-1</sup> )
Orange	10	125-150	1	2226
Red	20	250-300	1.2	557
Yellow	20	500-600	1	70
Green	30	850-1000	1.025	24

**Table 1.** Microplastic polyethylene spheres (purchased from 'Cospheric') and the mass (g) spiked into the chamber, and their size ranges ( $\mu$ m), density (g cm<sup>-3</sup>), and concentration (L<sup>-1</sup>) in the chamber.

- 3.2.2 Quality controls and blanks. Prior to the introduction of the microplastic particles ("spikes"), preliminary procedural blanks (0.1 L; n = 5) were taken of the artificial seawater to determine background contamination associated with the sea ice chamber.
- 3.2.3 Experimental procedure and sampling protocol. Once the procedural blanks were taken, the microplastic "spike" was added to the tank and left to mix for 24 hours. The following sampling protocol was then followed for Freeze 1:

- Seawater samples were taken from both the surface water (top 2 cm) (10 ml, n = 3)
   and subsurface (at 80 cm depth) (0.1 L, n = 3) layer.
- The system was then cooled down to promote ice growth within the tank. Once ~15 cm of ice had formed (3 days freezing at -25 °C), another set of seawater surface (10 ml; n= 3) samples were taken by drilling a hole into the ice and sampling the water just beneath the ice using a custom glass sampling pipe. Subsurface (0.1 L; n= 3) seawater samples were taken using the same custom glass sampling pipe, approximately 80 cm below the surface of the ice.
- Sea ice cores (n= 12) were then taken during one sampling period.
- After a two-day melting period (melting at 30 °C), surface (10 ml, n= 3) and subsurface (0.1 L, n= 3) samples of the meltwater were taken to determine particle distribution post-melt.

After re-cooling the tank for two days (at 0 °C), the following sampling protocols were followed for Freeze 2:

- Surface (10 mL, n=3) and subsurface (0.1 L, n=3) seawater samples were taken prior to ice formation.
- A second 3-day freezing period was undertaken (at -30 °C) resulting in ice formation with a depth of ~ 15 cm. Seawater surface samples (10 ml, n = 3) were taken by drilling a hole into the ice and sampling the water just beneath the ice using a custom glass sampling pipe. Subsurface samples (0.1 L, n= 3) were taken using the same sampling pipe, approximately 80 cm below the surface of the ice.
- Sea ice cores were taken (n = 12) using the same sampling procedures as before.
- After a melting period of one day, when the ice had partially melted, surface and subsurface seawater samples were taken (surface: 10ml, n= 3; subsurface: 0.1 L, n= 3) and again several hours later (surface: 10 ml, n= 3; subsurface: 0.1 L, n= 3) to track particle distribution at different stages of ice melt.
- After another 24h of melting, samples were taken when a small amount of ice was still present (surface: 10ml, n= 3; subsurface: 0.1 L, n= 3), and then again (a few hours later) when all the ice had melted (surface: 10ml, n= 3; subsurface: 0.1 L, n= 3), again to show particle distribution at different stages of ice melt.

All seawater samples were stored in 500 ml amber glass bottles which were rinsed with deionised water before use. Sea ice samples were collected in Freeze 1 (n = 12) (average core depth: 13 cm) and Freeze 2 (n = 12) (average core depth: 13 cm) from across the tank area. Samples were subsequently covered in polyethylene sheets and stored in a freezer (-40 °C) until further processing. Sea ice samples were sectioned into horizontal layers (1–3 cm thick) using an electric band saw in a cold room (-25 °C) and bulked to produce 3 bulk ice samples for Freeze 1 and 3 bulk ice samples for Freeze 2. Any particles residing on the outside of the cores were not included in the analysis, as these were presumed to be contamination from the surface of the ice during the ice core extraction process.

	Event	Air
		temperature
		(°C)
Days 1-6	Cool down system	2.6
Days 6-8	Freeze 1	-2.1
Days 9-10	Thaw 1	-0.3
Days 10-12	Cool down system	0
Days 12-15	Freeze 2	-2
Days 16-18	Thaw 2	0

**Table 2.** The timeline (days) of experimental events with the average chamber air temperatures as an indicator of the ice conditions in the seawater tank including air temperature (°C).

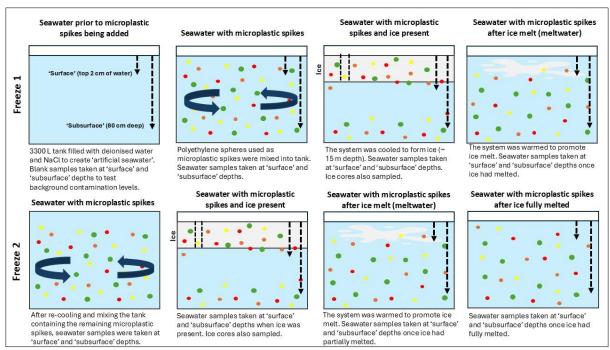


Figure 1. Diagram showing stages of experiment and sampling procedures.

3.2.4 Sample processing and analysis. Seawater and ice-melt samples were individually filtered through a 47 mm glass fibre filter paper. When filtering samples, deionised water was used to thoroughly rinse out the glass amber jars containing the sample and around the filtration funnel, ensuring all microplastics were transferred to the filter. Once the samples were on the filters, they were stored in labelled covered glass petri dishes until further analysis.

All filters were photographed (Canon EOS 1200D), both with and without fluorescence, and with a 5 mm × 5 mm grid overlay to aid manual counting. Due to the accumulation of very high numbers of spike particles on some of the filters (e.g. Layer 1 from all the ice cores), some filters were resuspended and were re-filtered through 2 or 3 filters to facilitate particle separation and counting. The particle counts from these additional filters were then combined to give the overall particle number for that sample.

3.2.5 Particle size and density. To understand whether the size or density of the particles influenced how the microplastic spikes were incorporated into the ice, the first set of replicate samples for surface and subsurface waters from both freeze experiments were manually counted using colour to discriminate between particle types. This was also undertaken for the ice cores from both freeze experiments, although ice layer 1 from both cores was excluded from this approach as there were too many particles accumulated on the filters to count

manually. Manual counting of samples was undertaken using images of sample filters with a grid overlay. These were uploaded into Image J (v0.5.8) and the counting tool was used to tally each coloured set of microplastic spikes, grid by grid.

- 3.2.6 Microplastic quantification. All filters were counted in ImageJ software using the 'Max Entropy' method as detailed in Geilfus et al (2019). Briefly, this entailed:
  - 1. Setting the scale of the picture
  - 2. Cropping the picture to focus on the filter
  - 3. Converting the image to 8-bit
  - 4. Adjusting the brightness and contrast level to optimise the visualisation of the particles.
  - 5. Applying a threshold following the maximum entropy ("Max Entropy") algorithm utilised in ImageJ
  - 6. Quantifying particles based on the area analysed (from the size detection limit to  $\infty$ ).

The Max Entropy threshold maximises the inter-class entropy (a measure of the uncertainty of an event taking place) (Geilfus et al., 2019) and works by automatically choosing the threshold that most effectively separates particles from the background, based on how much useful information each possible threshold would retain.

3.2.7 Enrichment Factor (EF) for microplastics in sea ice. An enrichment factor for microplastics in the upper layer of ice relative to surface water was calculated using the following equation. The average microplastic number concentration from both freeze experiments in the uppermost layer of ice was divided by the average microplastics number concentration from both freeze experiments in surface water. See Table S4 in Supporting Information for values.

$$EF_{MP} = \frac{C_{ice}}{C_{water}}$$

# 3.3 Results

3.3.1 Quality controls and accounting for spike particles in the sea ice chamber. The analysis of seawater blanks taken prior to the introduction of the Cospheric plastic spikes revealed very low levels of microplastics, containing mostly fibres of unknown polymer type. The likely

source of these fibres was from the chamber laboratory air but also through the use of cotton and polyester dominated surface-wipes used to clean the glass seawater tank prior to the addition of water. The very low levels of this debris (average particle number concentration: 42 particles L<sup>-1</sup>) were not considered high enough to interfere with the spike-particle transfer between ice and seawater during the experiments. Furthermore, the spike plastics were brightly coloured, engineered spheres which were readily observed and highly distinct from any other particulate material collected on the filters during the course of the experiments.

A summary of the spike plastic masses, and equivalent approximate particle numbers added to the chamber seawater prior to the freeze experiments are reported in Table 1 for each coloured plastic type. An attempt was made to account for particle movement (water to ice and ice to water) and their fate in the chamber over the course of the experimental period. Observations of particle movement during the experimental period revealed an important phenomenon whereby a fraction of each of the coloured spheres was deposited to the floor of the seawater chamber despite polyethylene densities being less than or similar to the density of the seawater. Once deposited, however, these particles were considered to be lost from the system as water circulation pumps situated near the bottom of the tank did not appreciably re-suspend these particles as the experimental period progressed. Deposition of the plastic particles to the chamber floor made a particle mass or number balance difficult to achieve (i.e. accounting for the number or mass added to the chamber prior to the experiments compared to those present in the sea water at the end of the experimental period). Besides particles being removed with successive seawater and ice samples, other loss processes (alongside deposition to the chamber floor) were considered negligible. An attempt was made to quantify the amount of particle spikes going in compared to the number of particles removed, by multiplying the average particle number concentration in seawater samples taken on the first day of the experiment (1.28 x 10<sup>4</sup>), by the chamber volume (3300 L). This resulted in a factor of 4 discrepancy, as the number of particles in was 9.18 x 10<sup>6</sup> (based on the number of spheres per gram for polyethylene provided by Cospheric), and the number of particles out was  $4.21 \times 10^7$ .

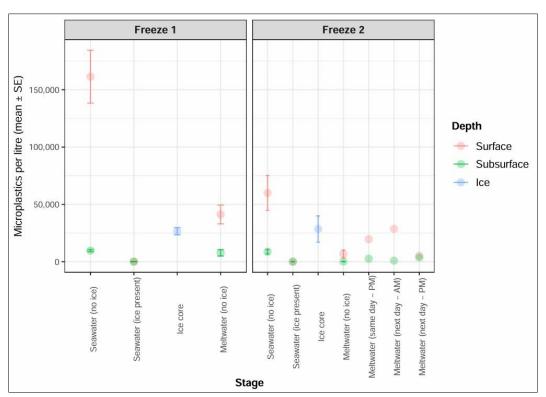
3.3.2 Visual observations of particle size and density in seawater. For all samples (surface and subsurface seawater and ice cores), the smallest orange polyethylene spheres (125 -150  $\mu$ m, 1 g cm) dominated out of the various microplastic spheres used in this experiment. For surface

water samples, the orange spheres made up 94 % of the total number of plastic particles, and for subsurface, 98 %. In the ice cores from Freeze-1 and Freeze-2, the orange spheres made up 92%, and 99.5 % of all the plastic particles respectively. All filters were also visually inspected to identify major differences in the colour and hence type of particles observed. The majority of filters containing particles corresponded to the amounts of each particle spiked into the tank; with orange being the most abundant, followed by red, then a small number of yellow particles, and very few of the largest green particles.

### 3.3.3 Observations of particle behaviour in seawater.

Stage	Freeze	Average particle concentration
		(L <sup>-1</sup> ) ± SD
Seawater	1	12769 ± 1772
Seawater with ice	1	52 ± 26
present		
Meltwater	1	8442 ± 4603
Seawater	2	9754 ± 3425
Seawater with ice	2	23 ± 1
present		
Meltwater	2	299 ± 262
Meltwater	2	4587 ± 1792
Meltwater	2	2833 ± 1902
Meltwater	2	4236 ± 2587

**Table 3.** The average particle concentration  $(L^{-1}) \pm SD$ , for seawater (Seawater = prior to ice formation, Ice = seawater with ice present, Meltwater = once ice had melted) in Freeze 1 and Freeze 2.



**Figure 2.** Surface (2 cm depth), Subsurface (80 cm depth) and Ice core microplastic number concentrations (L<sup>-1</sup>) for each stage of the experiment, split by Freeze 1 and Freeze 2.

The particle concentrations in the surface layer of the seawater (top ~2 cm) exhibited the same behaviour between the Freeze 1 and Freeze 2 experiments. A high concentration of particles was present in seawater prior to any ice, followed by a marked decrease in concentration once the ice had formed, followed by a slight increase in particles once the ice had melted.

For the subsurface seawater (80 cm below surface) the pattern of particle behaviour is similar to the surface water, whereby the concentration of particles in the deeper water declines markedly when ice is present. During the Freeze 1 experiment the particle number in the subsurface water increases following ice melt (similar to the surface water) but this increase is not apparent during the Freeze 2 experiment. Besides the variation marked by the replicate samples, the most likely explanation for this is the loss of particles from the system due to sinking and deposition to the floor of the chamber, and this is evident in both the surface and deeper waters during the Freeze 2 experiment. In Freeze 2, it is noteworthy that the particle concentration (average of the replicates) is lower than Freeze 1, due to removal of plastic particles from the chamber from repeated sample removal (ice cores and water) as well as loss of particles over the course of the experiments through deposition to the chamber floor.

Ice formation led to an almost complete absence of microplastic particles from the surface seawater (99.9 % decrease in surface microplastic particle number concentration within the seawater in Freeze 1 and Freeze 2). Similarly, Ice formation led to 99.5 % decrease in subsurface microplastics concentration within the seawater in Freeze 1, and a 99.8 % reduction in Freeze 2 (based on an average count of 3 replicates).

A 67 % reduction was seen in the microplastic concentration in surface seawater in between Freeze 1 and Freeze 2, and a 49 % reduction was seen in the microplastics concentration for subsurface seawater. We are assuming this loss is from seawater and ice core samples removed from the chamber, and some particles falling out of suspension to the bottom of the chamber.

3.3.4 Observations of particle behaviour in sea ice. 'Pancake ice' is commonly observed on seawater surfaces during initial ice formation in cold regions (Thomson et al., 2019) and was observed in the sea ice chamber in both freeze experiments. Interestingly, the buoyant polyethylene microplastic spheres used in this experiment were observed on the surface of the ice pancakes (Fig. 3).

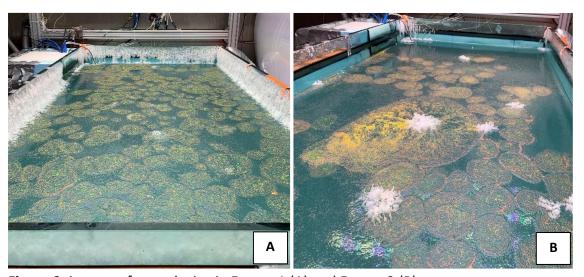


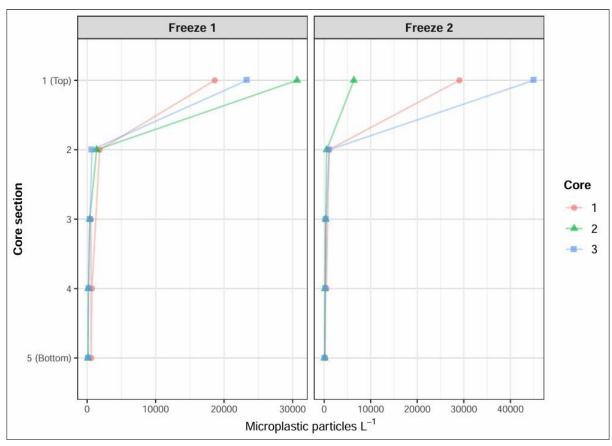
Figure 3. images of pancake ice in Freeze 1 (A) and Freeze 2 (B).

Freeze	Average particle
	concentration (L <sup>-1</sup> ) ± SD

1	3191 ± 747
2	2693 ± 1699

**Table 4.** Average particle concentrations  $(L^{-1}) \pm SD$ , for ice cores from Freeze 1 and Freeze 2.

In both Freeze 1 and Freeze 2, the ice-atmosphere layer (L1) contained the highest abundance of microplastics (Fig. 4), suggesting ice accumulates particles relative to the underlying seawater as indicated by the calculated enrichment factor ( $EF_{MP} = 12$ ). The number of microplastics then decreased through L2 - L5. Freeze 2 had a higher abundance of microplastics in L1 compared to Freeze 1 in 2 out of the 3 bulked cores. This is likely due to a higher presence of microplastic on the surface of the tank from the melting of Freeze 1, which resulted in a mass release of microplastic which then remained at the surface.



**Figure 4.** Vertical distribution microplastic particle number concentration in bulked ice cores (n = 3) from Freeze 1 vs. Freeze 2 by core section (1: ice - atmosphere interface, 5: ice - ocean interface). Average core length was 13 cm, and each layer was 2.5 cm thick.

3.3.5 Observations of particle behaviour in meltwater. Multiple meltwater samples were taken at the end of freeze 2 to track particle fate (Fig. 2). For surface seawater samples, particle

number concentrations continued to increase a few hours after the initial meltwater sample. This number generally decreased slightly the following day and then plateaued later in the day. For subsurface samples, a significant increase can be seen in particle numbers a few hours after the initial melt, which generally decrease the following day, and increases again later that same day. Evidence of particle accumulation in a buoyant, lower-saline (8 g  $^{-1}$ ) meltwater layer beneath the ice was seen during both melting stages for freeze 1 and 2. Most particles observed in this layer were the green polyethylene spheres (850 – 1000  $\mu$ m, 1.025 g cm<sup>-3</sup>).

### 3.4 Discussion

The aim of this study was to understand microplastic uptake into forming and fresh sea ice from sea water to see whether uptake into ice actually occurs, and to track particle fate during simulated seasonal thaw. Due to the low-density and small size of the particles, we hypothesised that the highest concentration of particles (particularly of the smallest diameter particles) would be found in the uppermost layer of sea ice. Some of the most common polymers found in the Arctic include polystyrene, polyethylene, polyester and polypropylene (Bergmann et al., 2017, Bergmann et al., 2019, Kanhai et al., 2019, Peeken et al., 2018), all of which are generally low-density polymers. As such, microplastic particles comprising of these polymer types are likely to exhibit neutral or positive buoyancy in seawater, although polymer additives and microbial/biological contamination (e.g. biofilm) may alter this buoyancy. The polymer of the microspheres used for this experiment was polyethylene, which is ubiquitous in the environment and provides insight into how low-density polymers might behave in real sea ice.

Several factors influence the likelihood of microplastics becoming entrapped in sea ice, including the concentration of microplastics in the water, the timing of ice formation, and the specific freezing conditions. High concentrations of microplastic pollution have been observed in Arctic seawater. In the Fram Strait, microplastics concentrations in the water column ranged between 0 and 1.287 particles  $L^{-1}$  (Tekman et al., 2020). In the same region, plastic debris was found in water samples at a concentration of 2.4 x  $10^{-3}$  ( $\pm$  8.0 x  $10^{-4}$ ) items  $L^{-1}$  (Morgana et al., 2018). In the Central Arctic Basin, surface waters underlying ice floes had microplastic concentrations that ranged from ND - 0.018 particles  $L^{-1}$ . Microplastics

concentrations in these samples were orders of magnitude lower than microplastic concentrations in sea ice cores (2 – 17 particles  $L^{-1}$ ), which further supports the theory that surface particles are readily incorporated into sea ice (Kanhai et al., 2020). In a study by Peeken et al (2018), extremely high concentrations of microplastics were found in ice sampled from the Fram Strait and Central Arctic. The highest particle number concentration (1.2 x  $10^4$  ( $\pm 1.4 \times 10^4$ )  $L^{-1}$ ) was found in an ice core taken from the older pack ice of the Fram Strait.

Although the ice chamber provides a controlled system for isolating the physical processes determining microplastic incorporation into sea ice and subsequent release in ice melt, several differences from real Arctic conditions should be acknowledged. The artificial seawater used in the experiment consisted solely of a simple salt solution and therefore lacked the complex mixture of dissolved minerals and biological material present in natural seawater, all of which can influence particle behaviour through processes such as aggregation, biofouling, and changes in effective density. In addition, the chamber environment does not replicate the turbulence, wind forcing, variable cooling rates, or biological activity that characterise sea ice formation in the field. The microplastic spikes used were also more uniform in size and shape than the heterogeneous particles found in the environment, with fibres being most prominent. As a result, the findings should be viewed as mechanistic insights rather than direct replicas of environmental behaviour.

Analysis of seawater and ice core samples from this study revealed that ice formation does result in the entrainment of plastic particles from the surrounding seawater (Fig. 2), and that the highest concentration of plastic particles reside in the uppermost layer of the ice (ice-atmosphere interface) (Fig. 4).

3.4.1 Observations of particle behaviour in seawater. The density of the particles ranged from  $1-1.2~{\rm g~cm^{-3}}$ . The density of the seawater was  $1.025~{\rm g~cm^{-3}}$  (based on an initial temperature of 20 °C and salinity of 35 g L<sup>-1</sup>). Despite efforts to keep the particles well mixed within the seawater, most of the denser red polyethylene spikes (density  $1.2~{\rm g~cm^{-3}}$ ) dropped out of suspension and sank, gathering around piping and equipment at the bottom of the chamber. Prior to ice formation, the microplastic spike particles mostly accumulated at the surface of the seawater in high concentrations (Fig. 2). For subsurface water samples prior to ice formation, microplastic particle number concentrations were also high, although approximately an order of magnitude lower than the surface. Upon visual inspection of the

filters from both Freeze 1 and 2, the orange particles (smallest, 125-150  $\mu$ m) were most abundant in both surface and subsurface samples, with some red particles (250-300  $\mu$ m) present and a small number of yellow particles (500-600  $\mu$ m), and very few green particles (largest, 850-1000  $\mu$ m). It is interesting to observe that despite a large number of red particles sinking in the chamber presumably due to their high density, a large number were still present in the surface water and water column in general.

Ice formation led to a 99.9% decrease in the microplastic number concentration in the surface water (beneath the ice) in both freeze experiments, clearly demonstrating that the microplastics were incorporated into the forming sea ice. Fluorescent images showed the remaining particles that were present in the surface water mostly consisted of the orange polyethylene spheres, one of the lowest density particles (1 g cm<sup>-3</sup>). This isn't something we would necessarily expect to see as these low-density particles were found in high concentrations in the surface layer of ice. It is possible that due to the abundance of particles in the surface water during ice formation, and the rapid rate of ice formation, that the ice reached a capacity for the number of particles that could be entrained, as seen in a simulation created by Mountford and Maqueda (2021). The orange particles were most abundant in the chamber; therefore, it seems plausible that these would be the particles leftover. Another possibility is that the orange particles were expelled in brine droplets during the initial stages of ice formation, when the ice crystals expel salt back into the water (NSIDC, 2024).

The enrichment factor for microplastics in sea ice indicates a significant enrichment of microplastics from the surface layer of seawater to the upper layer of ice core. This suggests that low density particles (with neutral or positive buoyancy in seawater) are readily entrained into new and growing sea ice and appear to have a vertical distribution in the ice that is similar to the distribution of ice brine. This, along with potential atmospheric deposition of microplastics from snow which has been highlighted as an important transport pathway (Bergmann et al., 2019), could increase the albedo effect and change both the permeability of sea ice and the absorption of solar radiation, also impacting sea ice melting (Peeken et al., 2018, Geilfus et al., 2019).

3.4.2 Observations of particle behaviour in ice. The highest concentration of microplastic particles were observed in the uppermost layer of the ice. Both cores from Freeze 1 and Freeze 2 demonstrated similar patterns, with the top layer of ice (Layer 1) containing the

highest concentrations of microplastics for all the ice cores taken in both freeze experiments (Fig. 4). An enrichment factor of 12 was calculated, further demonstrating that microplastics were concentrated in the ice at levels substantially higher than those present in the source seawater, indicating efficient particle incorporation during freezing. This enrichment is consistent with physical processes such as brine rejection and ice crystal growth that selectively retain low-density particles. In an environmental context, this level of enrichment supports the role of sea ice as a temporary sink that can subsequently release elevated microplastic loads during melt.

The number of particles was so high in the upper layer of ice that filters had to be resuspended and spread across multiple filters to enable counting (see Methods). Nonetheless, despite the high particle counts, the particle concentration varied between cores (some cores containing significantly more particles than others) which most likely reflects the heterogenous distribution of the particles observed across the surface of the water during initial freezing and the formation of pancake ice. Layer 2 of the ice cores from both freeze experiments possessed significantly fewer particle numbers compared to Layer 1. For example, there was a 94% decrease in particle numbers from L1 to L2 during Freeze 1, and a 95% decrease in particle numbers from L1 to L2 during Freeze 2. Particle numbers continued to decrease in layers 3, 4 and 5.

The vertical distribution of the particle concentrations in the ice does resemble the profile commonly observed for salinity, whereby the vertical salt profile in sea ice takes the form of a C shape: that is a high concentration at the top of the core followed by a decrease within the middle, and an increase again at the bottom. This has also been observed for other contaminant solutes like persistent organic pollutants, where results suggested that their incorporation into forming sea ice was influenced by brine advection (Garnett et al., 2019). It was therefore plausible that small particles like microplastics could demonstrate similar behaviour and be influenced by brine dynamics within young ice. However, for microplastics in sea ice we did not observe the increase at the bottom of the core (in closest proximity to the seawater), suggesting their incorporation and distribution in sea ice is less influenced by brine advection than dissolved solutes. This was also seen for the vertical distribution profiles of suspected microplastics in sea ice during a mesocosm experiment (Geilfus et al., 2019). Low, medium, and high concentrations of microplastics were spiked into separate concrete

pools which were left to freeze. Each profile showed an almost identical distribution to the ones in this study, with the highest number of particles found in the upper layer of ice and very few particles found in the rest of the layers, and no increase at the bottom layer. Most particles were in the size category 125 - 63  $\mu$ m and 250 - 125  $\mu$ m, which was also seen in this study, with the smaller orange particles (125 -150  $\mu$ m) being the most abundant in most samples.

Pancake ice formed at the surface of the chamber seawater in both freeze experiments (Fig. 3). This resulted in conglomerations of particles across the tank, which is likely the reason some cores contained higher concentrations of microplastics at the surface than others. The pancakes could be seen forming before the water was fully frozen, possibly as ice crystals were forming and aggregating (commonly referred to as frazil ice in Polar marine environments), and influencing the behaviour of the buoyant microplastic particles. Pancake ice in the Arctic is formed due to increased wave activity (Nose et al., 2021). In this experiment, this is most likely explained by the pump used to keep the seawater well mixed.

3.4.3 Observations of particle behaviour in meltwater. Microplastic release during simulated sea ice thaw was also tracked, with visible evidence of particle accumulation in a buoyant, lower-saline meltwater layer beneath the ice. This could result in a high concentration of particles in a relatively thin layer of water adjacent to the underside of the ice. Visually, it was apparent that the larger green particles were present in this beneath ice water layer, although visual analysis of the surface water samples revealed that the particle number concentration was dominated by the smaller orange particles.

Meltwater samples revealed a 99.7% increase in microplastic number concentration within the surface water during Freeze 1, and 98.9% increase in Freeze 2. For subsurface water, there was a 99.4% increase in freeze 1, and an 87.1% increase in freeze 2. In freeze 2, the number of particles overall was lower due to overall loss of particles from the system (i.e. deposition to the chamber floor) and the accumulative loss through repeated removal of water and icecore samples. In the Arctic, this mass release of particles could have a significant negative impact on sympagic organisms. The ingestion of microplastics by these organisms can lead to various adverse effects, including physical harm, reduced feeding efficiency, and potential transfer of pollutants associated with microplastics (Cole et al., 2013). Ingestion can also lead

to microplastics bioaccumulating through the food web (Miller et al., 2020) or inadvertently being transported to the deeper ocean.

#### 3.5 Conclusion

The results from this study provide further evidence that sea ice is a significant repository and transport mechanism for microplastics. In the Arctic, release of microplastics during ice melt with seasonal thaw will be complicated by numerous factors, including the presence of an ice rafted snowpack, the likely presence of melt ponds and the overall rate of ice thawing. Nonetheless, ice thaw is likely to play an important role in the geochemical cycling of microplastics in Arctic surface waters and their seasonal release into a beneath ice water layer could have significant negative effects on the environment, affecting both ecosystems and the organisms within them. Marine organisms, ranging from microscopic plankton to larger marine animals, can be exposed to these pollutants. Further research is required to assess the potential impacts of microplastics on Arctic marine biota, particularly ice-associated organisms.

# Acknowledgements

The work in this chapter was a collaborative effort with the team at the Roland von Glasow Air-Sea-Ice Chamber (RvG-ASIC) at the University of East Anglia. I would particularly like to thank Jan Kaiser and Nick Garrard from UEA for their help during this work, and Andrew Meas for supplying us with photography equipment. A big thank you also goes to Jack Garnett for his help during this experiment. Although we argued like siblings, his knowledge and experience was invaluable, and our coffee breaks and post-work debriefs kept me sane(ish).

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# **Supporting Information**

Table S1: particle number concentration (L<sup>-1</sup>) in blank samples.

Blank	Sample	MP	Particle
sample	size (g)	count	L <sup>-1</sup>
1	109.02	10	92
2	94.8	5	53
3	106.4	5	47
4	101	0	0
5	101.3	2	20

Table S2: Sampling information and particle number concentration (L<sup>-1</sup>) for seawater in both freeze experiments.

Replica te	Freez e	Environm ent	Depth	Sample code	MP count	Samp le size (g)	Particle number concentrati on (L <sup>-1</sup> )
1	1	Seawater	Surface	14-09-21 MP SW (Surface) S1	1295	7.56	176092.6
2	1	Seawater	Surface	14-09-21 MP SW (Surface) S2	1290	11.42	116122.6
3	1	Seawater	Surface	14-09-21 MP SW (Surface) S3	1690	9.05	191969.1
1	1	Seawater	Subsurfa ce	14-09-21 MP SW S1	977	102.2 5	9822.553
2	1	Seawater	Subsurfa ce	14-09-21 MP SW S2	1125	102.3 1	11303.88
3	1	Seawater	Subsurfa ce	14-09-21 MP SW S3	767	97.58	8080.303
1	1	Seawater with ice present	Surface	19-09-21 MP SW (surface) S1	5	96.04	53.51937
2	1	Seawater with ice present	Surface	19-09-21 MP SW (surface) S2	24	99.51	247.9349
3	1	Seawater with ice present	Surface	19-09-21 MP SW (surface) S3	11	102.4 8	110.3435
1	1	Seawater with ice present	Subsurfa ce	19-09-21 MP SW S1	7	103.1 7	69.74896
2	1	Seawater with ice present	Subsurfa ce	19-09-21 MP SW S2	2	99.51	20.66124

3	1	Seawater	Subsurfa	19-09-21 MP SW S3	6	102.8	59.99416
5	1	with ice	ce	15 05 21 1011 500 55		1	33.33410
		present				-	
1	1	Meltwater	Surface	20-09-21 MP SW	261	6.97	38494.69
1	1	Meitwater	Surface	(surface) S1	201	0.97	36494.09
2	1	0.4 - 14 4	Conford	, ,	405	0.07	F C 7 2 0 4
2	1	Meltwater	Surface	20-09-21 MP SW	495	8.97	56729.1
_				(surface) S2			
3	1	Meltwater	Surface	20-09-21 MP SW	424	15.24	28600.52
				(surface) S3			
1	1	Meltwater	Subsurfa ce	20-09-21 MP SW S1	862	98.66	8981.715
2	1	Meltwater	Subsurfa	20-09-21 MP SW S2	1151	100.8	11737.21
۷	+	ivicitwatei	ce	20-09-21 WIF 3W 32	1131	1	11/3/.21
3	1	Moltwator	Subsurfa	20 00 21 MD CM/C2	222	ļ <del>-</del>	2500 270
3	1	Meltwater	ce	20-09-21 MP SW S3	232	91.79	2598.279
1	2	Seawater	Surface	22-09-21 MP SW	529	14.99	36278.32
_				(surface) S1			
2	2	Seawater	Surface	22-09-21 MP SW	1116	12.96	88522.22
_	-	Scawater	Janace	(surface) S2	1110	12.50	00322.22
3	2	Seawater	Surface	22-09-21 MP SW	639	11.89	55247.43
3	2	Seawatei	Juliace	(surface) S3	039	11.03	33247.43
1	2	Convetor	Cubcurfo	•	712	00.20	7292 706
1	2	Seawater	Subsurfa ce	22-09-21 MP SW S1	713	99.28	7382.796
2	2	Seawater	Subsurfa	22-09-21 MP SW S2	613	102.4	6149.741
			ce			7	
3	2	Seawater	Subsurfa	22-09-21 MP SW S3	1249	101.4	12651.22
J	-	Jean acc.	ce	22 03 22 1111 311 03	12.13	9	12001.22
1	2	Seawater	Surface	26-09-21 MP SW	1	12.08	85.09934
_	-	with ice	34.1466	(surface) S1	-	12.00	03.0333 .
		present		(Surface) Si			
2	2	Seawater	Surface	26-09-21 MP SW	1	14.62	70.31464
۷		with ice	Juliace	(surface) S2	*	14.02	70.51404
		present		(Surface) 32			
3	2	Seawater	Surface	26-09-21 MP SW	1	13.09	78.53323
J	-	with ice	Juliace	(surface) S3	1	13.09	/0.33323
				(Surface) 33			
1	1	present	Cubacet	26 00 24 MD CM C4	12	07.74	21.04100
1	2	Seawater	Subsurfa	26-09-21 MP SW S1	2	97.71	21.04186
		with ice	ce				
_		present	6.1.6	26.00.04.4.7.000.05		00.00	20 50525
2	2	Seawater	Subsurfa	26-09-21 MP SW S2	2	99.82	20.59707
		with ice	ce				
		present					
3	2	Seawater	Subsurfa	26-09-21 MP SW S3	2	90.63	22.68564
		with ice	ce				
		present					
1	2	Meltwater	Surface	27-09-21 MP SW	139	11.93	11977.54
				(surface) S1 AM			
2	2	Meltwater	Surface	27-09-21 MP SW	76	9.61	8129.865
				(surface) S2 AM			
3	2	Meltwater	Surface	27-09-21 MP SW	3	10.99	280.6187
)	_	· · · · · · · · · · · · · · · · · · ·	Januace	(surface) S3 AM		10.55	200.0107
			I	(Juliuce) JJ Alvi	_1	ı	L

Meltwater	Subsurfa			102.6	440.4713
	ce	27-09-21 MP SW S1 AM	44	9	11011723
Meltwater	Subsurfa	27-09-21 MP SW S2	1	103.0	9.976708
Meltwater			5	_	49.85935
Meltwater			132		16290.04
Meltwater	Surface	27-09-21 MP SW	143	13.95	10537.92
		(surface) S2 PM			
Meltwater	Surface	27-09-21 MP SW	225	11.77	19651.66
		(surface) S3 PM			
Meltwater	Subsurfa	27-09-21 MP SW S1	399	98.9	4147.341
	ce	PM			
Meltwater	Subsurfa	27-09-21 MP SW S2	660	107.8	6290.376
	ce	PM		6	
Meltwater	Subsurfa	27-09-21 MP SW S3	239	92.47	2656.991
	ce	PM			
Meltwater	Surface	28-09-21 MP SW	97	12.2	8173.443
		(surface) S1 AM			
Meltwater	Surface		113	7.59	15304.87
Meltwater	Surface		347	12.45	28651.89
		1			
Meltwater			465		4685.552
Meltwater			185	96.71	1966.498
				0= 0=	0== 0004
Meltwater			91	97.87	955.8394
NA altropatan	1		100	11.61	14002.40
Meitwater	Surrace		199	14.61	14002.19
Moltwator	Curfoco	·	166	10.07	0042.240
Meitwater	Surface		100	10.07	9043.349
Moltwator	Curface	<u>'</u>	77	15 17	5217.93
ivieitwater	Juliace		' '	13.1/	3217.93
Meltwater	Subcurfa	·	686	101.2	6965.705
ivicitwatei			000		0505.705
Meltwater			181	_	1746.461
IVICIEWALEI			101		1,40.401
Meltwater	+		392	+	3679.81
ivicitivatei	ce	PM	332	105.5	30,3.01
	Meltwater  Meltwater  Meltwater  Meltwater  Meltwater  Meltwater  Meltwater  Meltwater	Meltwater Subsurface  Meltwater Surface  Meltwater Surface  Meltwater Subsurface  Meltwater Subsurface  Meltwater Subsurface  Meltwater Subsurface  Meltwater Surface  Meltwater Surface  Meltwater Surface  Meltwater Surface  Meltwater Surface  Meltwater Subsurface  Meltwater Surface  Meltwater Surface  Meltwater Surface  Meltwater Surface  Meltwater Subsurface  Meltwater Subsurface	Ce	Meltwater         ce         AM           Meltwater         Subsurfa ce         27-09-21 MP SW S3 ce         5           Meltwater         Surface         27-09-21 MP SW (surface) S1 PM         132           Meltwater         Surface         27-09-21 MP SW (surface) S2 PM         143           Meltwater         Surface         27-09-21 MP SW S2 (surface) S3 PM         225           Meltwater         Subsurfa ce         27-09-21 MP SW S1 S2 PM         399           Meltwater         Subsurfa ce         27-09-21 MP SW S2 PM         660           Meltwater         Subsurfa ce         27-09-21 MP SW S3 Ce         239           Meltwater         Subsurfa ce         28-09-21 MP SW S3 Ce         239           Meltwater         Surface         28-09-21 MP SW S3 Ce         347           Meltwater         Surface         28-09-21 MP SW S1 Ce         347           Meltwater         Subsurfa ce         28-09-21 MP SW S1 Ce         465           Meltwater         Subsurfa ce         28-09-21 MP SW S1 Ce         465           Meltwater         Subsurfa ce         28-09-21 MP SW S3 Ce         91           Meltwater         Surface         28-09-21 MP SW S3 Ce         91           Meltwater         Surface         2	ce         AM         4           Meltwater         Subsurfa ce         27-09-21 MP SW S3 and Surface         5         103.0 g           Meltwater         Surface         27-09-21 MP SW (surface) S1 PM         132         8.33           Meltwater         Surface         27-09-21 MP SW (surface) S2 PM         143         13.95           Meltwater         Surface         27-09-21 MP SW (surface) S3 PM         225         11.77           Meltwater         Subsurfa ce         27-09-21 MP SW S1         399         98.9           Meltwater         Subsurfa ce         27-09-21 MP SW S2         660         107.8           Meltwater         Subsurfa ce         27-09-21 MP SW S3         239         92.47           Meltwater         Subsurfa ce         28-09-21 MP SW S3         239         92.47           Meltwater         Surface         28-09-21 MP SW S3         239         92.47           Meltwater         Surface         28-09-21 MP SW         113         7.59           Meltwater         Surface         28-09-21 MP SW         113         7.59           Meltwater         Subsurfa ce         28-09-21 MP SW S3         465         102.0           AM         AM         28-09-21 MP SW S3         91

Table S3: Sampling information and particle number concentration (L<sup>-1</sup>) for ice cores in both freeze experiments.

Freeze	Core	Layer	MP count	Sample size (g)	Particle number concentration (L <sup>-1</sup> )
1	1	1	3507	190	18642
1	1	2	774	430	1818

1       1       1       4       264       380       702         1       1       5       224       390       580         1       2       1       6982       230       30660         1       2       1       6982       230       30660         1       2       2       629       460       1381         1       2       3       175       460       384         1       2       4       78       440       179         1       2       5       43       380       114         1       3       1       4622       200       23341         1       3       1       4622       200       23341         1       3       2       306       440       702         1       3       3       185       450       415         1       3       4       83       430       195         1       3       5       62       400       157         2       1       1       4359       150       29060         2       1       2       452       430						
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	2	3	2	491	450	1091
2 3 4 124 400 310	2	3	3	197	440	448
	2	3	4	124	400	310
2 3 5 58 450 129	2	3	5	58	450	129

Table S4: values used to calculate Enrichment Factor.

	Average Average				
	MP	MP			
	count in	count in			
	L1 of ice	surface			
	cores	water			
Freeze 1	24215	3228			
Freeze 2	26815	1200			
	51030	4428			
EF	$EF_{MP} = \frac{C_{ice}}{C_{water}}$				
	= 12				

# 4. Measuring the body burden of microplastics in Arctic Copepods from the Fram Strait.

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#### **Abstract**

Microplastics have been found in high concentrations in the Arctic in a wide range of samples, suggesting they are readily available to marine biota. The incorporation of microplastics in zooplankton represents a critical aspect of microplastic pollution in marine ecosystems. Zooplankton play a vital role in the biological carbon pump, transporting and sequestering carbon in the deep ocean. As microplastics are known to be ingested by zooplankton, it is possible that they are also transporting these particles to the deep ocean during their seasonal migration. This study investigates the concentration of microplastics in the copepod species Calanus finmarchicus, utilising samples collected during research cruises in May 2018 and August 2019 from the Fram Strait in the Arctic Ocean. Samples were collected from both surface and deep-water layers to determine whether microplastics were being transported by C. finmarchicus from surface to deep waters as they descend into diapause. Individuals in samples from surface waters contained microplastics exclusively during the May cruise, while microplastics were detected in individuals from both surface and deep waters in August. Results from this study suggest that the body burden of microplastics in *C. finmarchicus* is very low, and it is unlikely that they are transporting microplastics to the deep ocean in large numbers as they enter diapause, but in some cases, it is possible for microplastics to be retained for longer periods. This study emphasises the importance of investigating microplastic ingestion by key marine organisms and highlights the need for further research to understand seasonal dynamics and ecological implications.

#### 4.1 Introduction

Zooplankton play a vital role in the marine ecosystem, serving as a crucial link between primary producers and higher trophic levels (Jónasdóttir et al., 2015). In the Arctic, these tiny, drifting organisms include a diverse range of species, such as copepods, krill, and various larval forms of marine animals (Kosobokova and Hirche, 2009). Despite their small size, zooplankton collectively form massive swarms providing a significant food source for larger marine creatures like fish, seabirds, and marine mammals (Krumhansl et al., 2018). Zooplankton are also key players in nutrient and carbon cycling, transporting and sequestering carbon in the deep ocean which they do by feeding on the primary production (phytoplankton) in surface waters and producing sinking faecal pellets, by the deposition of dead zooplankton, or through the lipid pump, a process whereby zooplankton store carbon as fats in summer and then migrate to deep ocean waters in the winter (Jónasdóttir et al., 2015; Steinberg and Landry, 2017). The Arctic's extreme environmental conditions present unique challenges for zooplankton survival and reproduction. Nonetheless, these resilient organisms have adapted to thrive in this harsh environment, contributing to the overall health and productivity of the Arctic marine ecosystem (Gunaalan et al., 2023).

In the Arctic and sub-Arctic seas, *Calanus* spp. dominate the biomass of the mesozooplankton community by up to 75–90 % (Gabrielsen et al., 2012). The calanoid copepods *Calanus glacialis*, *Calanus finmarchicus* and *Calanus hyperboreus* are the dominant zooplankton taxa across the North Atlantic, Arctic shelf seas and the open Arctic Ocean (Melle et al., 2014). The planktonic copepod, *Calanus finmarchicus*, is one of the most important multicellular zooplankton species in the northern North Atlantic, based on its abundance and role in food webs and biogeochemical cycles (Melle et al., 2014). Because of their distribution in Arctic and Atlantic waters, *C. finmarchicus* are used as climate indicators (Gabrielson et al., 2012), making them an important component of marine life in the Arctic.

The copepod lifecycle (Fig. 1) typically involves the following stages: the non-feeding stages, which include eggs and nauplii, feeding and rapid growth which takes place from the later nauplii stages to copepodite stage C2, followed by the continued development to C5 (Anderson et al., 2022). During copepodite stages C3 to C5, digested food is increasingly used for lipid production, which forms the energy reserves needed to sustain the animals through diapause (Jónasdóttir et al., 2015). Diapause is characterised by a pause in development and

feeding, and reduced metabolic rates, which occur at fixed stages of development, typically only once during the life cycle of *Calanus finmarchicus* (Krumhansl et al., 2018). It occurs in the autumn, when development pauses at stage C5, and populations descend to depths of 600 - 1400 m into the mesopelagic zone below the mixed layer (Jónasdóttir et al., 2022, Skaret et al., 2014). In the spring, populations migrate back to the surface waters at stage C6, where they feed on the developing phytoplankton and develop into the adults that initiate the next generation (Jónasdóttir et al., 2015, Krumhansl et al., 2018). Microplastics can overlap in size with phytoplankton (Gunaalan et al., 2023), so it is possible that during their seasonal migration, when *C. finmarchicus* are feeding on the developing phytoplankton blooms in surface waters, microplastics could be inadvertently ingested and subsequently transported from surface waters to the deeper ocean (Fig. 1) (Vroom et al., 2017, Botterell et al., 2022, Gunaalan et al., 2023) through the rapid sinking of feacal pellets (Katija et al., 2017). Here, they can accumulate in deep-sea sediments or be ingested by benthic organisms leading to potential transfer to higher trophic levels (Rodríguez-Torres et al., 2020).

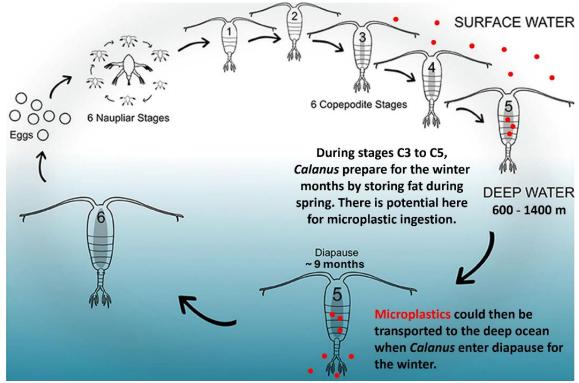


Figure 1. The life cycle of Calanus finmarchicus, modified from Mayor et al (2020).

As the gateway that transfers warm Atlantic water, via the West Spitsbergen Current to the Central Arctic, the Fram Strait plays a vital role in carrying anthropogenic pollutants to this remote part of the world (Peeken et al., 2018). High concentrations of microplastic have been

reported in samples from the Fram Strait, including in the water column (Tekman et al., 2020), snow (Bergmann et al., 2019), sea ice (Peeken et al., 2018), and more recently, ice algae (Bergmann et al., 2023), proving they are readily available to marine biota in the area. Microplastics can be incorporated by biota in a number of ways, such as filter feeding, suspension feeding, inhalation at air-water surface, consumption of prey exposed to microplastics, or via direct ingestion (Anderson et al., 2016). Multiple species of zooplankton in the Fram Strait have been found to ingest microplastic particles (Botterell et al., 2022). Once inside the animals, the plastic particles can disrupt physiological processes, impede feeding efficiency, and reduce energy reserves. The accumulation of microplastics in the digestive tract may lead to gut blockage, compromising nutrient absorption and posing a toxic risk. Some research suggests ingestion of microplastic can alter zooplankton behaviour, including reduced vertical migration and avoidance responses, which may ultimately alter trophic interactions within the food web (Vroom et al., 2017). Microplastic uptake by zooplankton is likely to increase as the Arctic continues to warm due to climate change. One of the impacts of warming Arctic summers and the subsequent reduction in sea ice cover, may be an increase in plastic debris in the Arctic (Mallory, 2008). Ice from the Fram Strait and Central Arctic has been reported to contain extremely high microplastic concentrations ((1.2  $\pm$  1.4)  $\times$  10<sup>4</sup> L<sup>-1</sup>) (Peeken et al., 2018).

The spring bloom in the open waters of eastern Fram Strait is thought to be initiated when surface heat fluxes act to stratify the water column during the month of May (Schourup-Kristensen et al., 2021), and peaks during early June (Engel et al., 2019). Cherkasheva et al (2014) found that, based on chlorophyll-a (CHL) concentrations, the start of the bloom in the eastern Fram Strait area generally occurred in May, and started as late as July-August in the western part of the Fram Strait. The Fram Strait is a complex physical environment, where variables such as sea ice cover and heat can affect the timings and distribution of primary production (Cherkasheva et al., 2014).

In this study, we investigate the body burden of microplastics in the marine copepod *Calanus finmarchicus*, planktonic filter feeders between 2 and 4 mm in length, in stage C5, to determine whether they are acting as a vector for microplastic particles from surface to deep waters as they descend into diapause. *C. finmarchicus* rise to the surface to feed on phytoplankton during the spring bloom and increase their lipid stores in preparation for

diapause. If they inadvertently ingest microplastics during this time, then we could assume they are transporting these microplastics down to the deep ocean when they descend for diapause. We might, therefore, expect to see a higher microplastic content in individuals sampled in surface waters during the May cruise, as this is when many *C. finmarchicus* will be feeding on the spring bloom, and higher microplastic content in deep samples from the August cruise as by this point, many *C. finmarchicus* will have increased their lipid stores ready for diapause.

#### 4.2 Materials & methods

4.2.1 Sample collection. Zooplankton samples were collected during two research cruises on the RRS James Clark Ross: May 2018 (JR17005) and August 2019 (JR18007), from both surface and deep waters (Fig. 2). Samples taken during the May cruise likely represent the population of *C. finmarchicus* feeding on the spring bloom, and those taken during the August cruise will include some *C. finmarchicus* still feeding on the bloom, but also some that have entered diapause. The surface samples were collected using a Bongo net (2 x 57 cm diameter rings and 200  $\mu$ m mesh spacing), and deep samples were collected using a MOCNESS multinet (1 m² mouth opening with 9 x 330  $\mu$ m meshed nets). The focus area of the cruises was the Fram Strait, where deep basins coincide with seasonal sea ice, creating ideal conditions for *Calanus* to accumulate large fat stores and enter diapause in deep ocean layers. *C. finmarchicus* were also sampled at two ice stations during the August 2019 cruise, using Bongo nets deployed from 200 - 0 m.

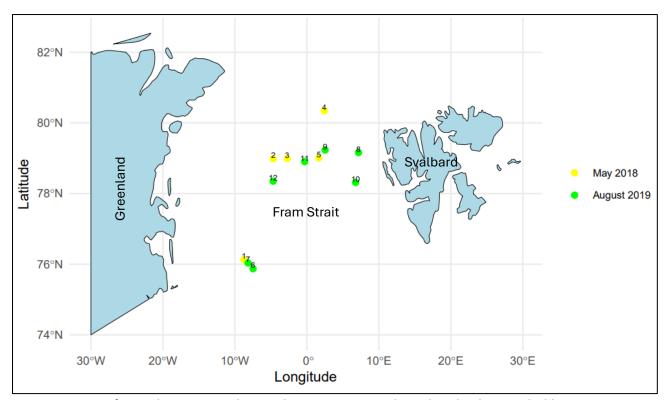


Figure 2. Map of sampling area with sampling stations numbered and colour coded by cruise.

4.2.2 Sample processing. Based on availability, a total of 22 composite samples were analysed (15 individuals per sample, n = 330). The first set of samples were taken during the May 2018 cruise, from both surface (n = 5) and deep (n = 5) water. The second set of samples were taken during the August 2019 cruise, from surface (n = 5) and deep (n = 5) water, and ice stations (n = 2).

4.2.2.1 Picking. Individual *C. finmarchicus* specimens (2 - 4 mm long) were manually extracted from larger bulk samples stored in formalin. These larger samples were filtered out of formalin and rinsed thoroughly with deionised water. The contents of the larger samples were transferred into a glass petri dish, which was then observed under a Leica (M50) microscope where *C. finmarchicus* were picked out and carefully rinsed with deionised water to ensure any plastic on the outside of the animal was removed, before being placed into a 5 ml glass tube containing formalin until further analysis.

4.2.2.2 Digestion. The method for digestion was modified from a study by Kallenbach et al (2021) which tested four protocols for dissolving organisms with a chitin exoskeleton for microplastic analysis and found that the use of  $H_2O_2$  followed by chitinase was a highly efficient method and revealed high recovery rates for microplastic fibres, beads and tyre

particles. Samples were filtered out of formalin onto stainless steel mesh filters (50  $\mu$ m pore size) using a Buchner funnel filtration system. From here, the individual animals were moved from the mesh into a 25 ml glass beaker using tweezers (15 animals per beaker). A shallow layer (5 ml) of 30 % aqueous  $H_2O_2$  solution was added to each beaker. The beakers were covered with foil and transferred into a shaking oven/incubator (SHEL LAB) and left for 24 hours at 50 °C. After 24 hours, the beakers were removed, and 2 ml chitinase (96 U L<sup>-1</sup>) was added straight into each beaker. These were once again left to digest at 37 °C for 48 hours. Some carapaces remained in the samples, which were carefully removed using tweezers, and rinsed gently with deionised water into the beaker to ensure all particles remained within the sample. The final samples were poured onto a clean stainless steel mesh filter using the filtration system to clean off digestion residue. Each filter was then carefully washed to transfer the sample off the mesh filter back into the beaker. This remaining sample was then carefully transferred onto silver filters using a glass pipette, ready for  $\mu$ FT-IR analysis.

4.2.3 Microplastic identification. Scanning  $\mu$ FT-IR (PerkinElmer Spotlight 400, UK) analysis was performed in reflectance mode at a pixel resolution of 8 cm<sup>-1</sup> with 4 scans per pixel. An interferometer speed of 2.2 cm s<sup>-1</sup> was used, and scans were carried out from 4000 to 750 cm<sup>-1</sup> and included particles down to 25  $\mu$ m. Each sample took 89 minutes to complete. A microplastic identification software (siMPle v1.0.0) was then used to determine microplastic number concentration, as well as the polymer types and sizes of the microplastics found in the samples. Each identified particle was assigned a correlation score, representing the similarity between the measured spectrum and the reference polymer. Only particles with correlation values  $\geq$  0.5 (or  $\geq$  50%) were accepted as confidently identified.

4.2.4 Contamination mitigation and blanks. Sample preparation and analysis was undertaken in an ISO-5 dedicated microplastic clean laboratory with controlled access and a HEPA filter. Except for µFTIR analysis, processing also took place within a laminar flow cabinet (class II micro-flow biological safety cabinet, which filters air through a 99.999% high-efficiency particulate air (HEPA) filter (MDH Contamination Control, Hitchings Clinical Services, UK).

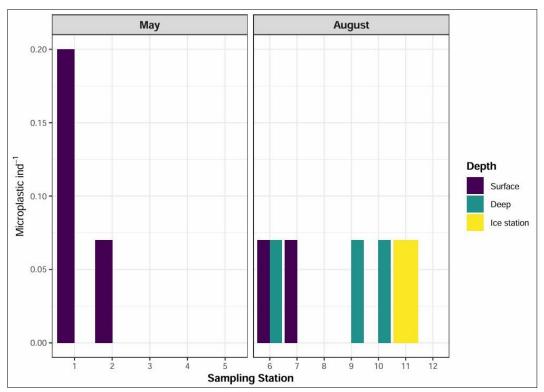
Blank samples (n = 4) were taken by running deionised water through the exact processing steps as the real samples. Artificially modified cellulose was found in 3 out of 4 blanks, and 4 environmental samples. No other polymers were present in the blank samples. Samples were adjusted to compensate for the contamination by artificially modified cellulase by subtracting

the average number of particles found in the blank samples from samples containing the same polymer. The average number of artificially modified cellulose particles was 1.

#### 4.3 Results

Over half of the samples contained microplastics, although the numbers were very low, with a maximum incidence of 0.2 particles per individual (3 particles per 15 pooled individuals) found in surface samples from the May 2018 cruise (Fig. 3).

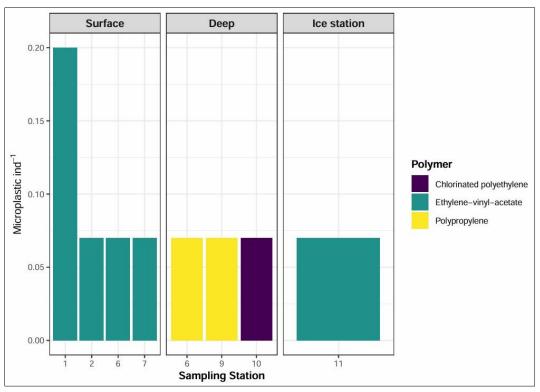
4.3.1 Microplastic findings in C. finmarchicus. Individuals in surface water samples from both cruises contained microplastics, whereas only individuals in deep water samples from the August 2019 cruise contained microplastics. A total of 0.07 particles per individual (1 particle per 15 pooled individuals) were found at the two ice stations.



**Figure 3.** Microplastic particle incidence in zooplankton (*C. finmarchicus*) samples from surface (n = 5), deep-water (n = 5) and ice stations (n = 2) samples, for both sampling cruises (May 2018 and August 2019).

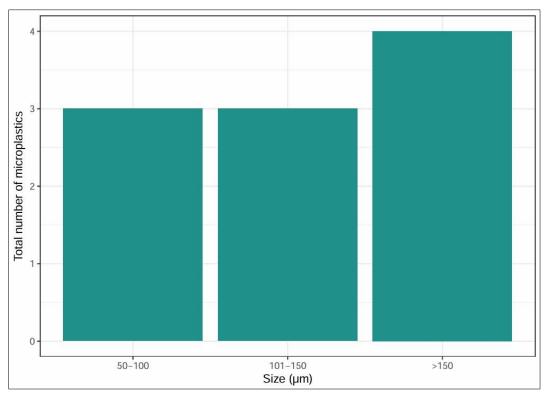
4.3.2 Microplastic polymer types. A clear difference can be seen in the type of polymers found in surface samples versus deep, with deep samples containing a mixture of chlorinated

polyethylene (PE-CI) and polypropylene (PP), and surface samples containing only ethylene-vinyl-acetate (EVA) (Fig. 4).



**Figure 4.** Microplastic particle incidence by polymer type in zooplankton (*C. finmarchicus*) samples from surface (n = 5), deep-water (n = 5) and ice station (n = 2) samples.

4.3.3 Microplastic particle size. Microplastic particles detected in *C. finmarchicus* samples were split into four size categories: <50, 50–100, 101–150, and >150  $\mu$ m. No microplastics <50  $\mu$ m were found in individuals, with most particles being between 101-150 or >150  $\mu$ m (Fig. 5).



**Figure 5.** Microplastic particle incidence split into three size categories: 50–100  $\mu$ m, 101–150  $\mu$ m and >150  $\mu$ m. The lower size cut-off was 25  $\mu$ m based on the pixel size selected for  $\mu$ FTIR analysis.

#### 4.4 Discussion

The purpose of this study was to investigate the incorporation of microplastics in species C. finmarchicus from both surface and deep-water samples, during two important months (May and August) in the Calanus life cycle, to determine whether microplastics are being transported from surface to deep waters as C5 C. finmarchicus enter diapause. C. finmarchicus was selected for this study due to its ecological importance as a dominant Arctic and sub-Arctic copepod and a key transport mechanism for energy transfer from primary producers to higher trophic levels, including fish, seabirds and marine mammals. Given its high feeding rates and vital role in marine food webs, substantial microplastic ingestion by this species raises concerns not only for individual health, but also for the potential transfer and amplification of microplastics and associated contaminants through Arctic marine ecosystems. Using  $\mu$ FT-IR analysis, this study confirms the presence of microplastics in zooplankton species C. finmarchicus, although concentrations were low.

There have been many laboratory studies investigating and experimentally confirming microplastic ingestion by zooplankton, however the concentrations of microplastics used in these studies are disproportionately high, usually orders of magnitude higher than what is currently reported in environmental samples. Findings from field studies show that microplastics are ingested by zooplankton in the environment, however the results are varying, and the studies are lacking.

There are few studies that have looked at microplastic ingestion by *Calanus* in environmental samples, with most studies choosing to investigate the effects of microplastic ingestion in controlled laboratory studies. However, Botterell et al (2022) looked at the ingestion of microplastics by a range of copepod species and found that microplastic ingestion in *C. finmarchicus* was  $0.01 \pm 0.003$  per individual, compared to  $0.21 \pm 0.03$  in *C. hyperboreus*. In the Northeast Pacific Ocean, 0.029 particles per *Calanus* individual was reported (Desforges et al., 2015), and in the Black Sea, microplastic ingestion of  $0.024 \pm 0.020$  particles per individual was found (Aytan et al., 2022). Despite the small sample size in this study, the results are therefore consistent with observations from other studies investigating microplastic concentrations in *C. finmarchicus*.

4.4.1 Microplastic concentrations in C. finmarchicus. Almost half of the stations sampled in this study contained individuals that had not consumed any microplastics. Based on the low numbers in pooled samples, we can assume that actually, the majority of individuals did not contain microplastics. Incorporation of microplastics in zooplankton is mostly determined by feeding habits (Xu et al., 2022, Gunaalan et al., 2023). However, one study observed copepods exhibiting a taste discrimination towards microplastics, rejecting 80 % of the microplastics after touching them with their mouth parts (Xu et al., 2022), so it is possible that low microplastic uptake in copepods is simply due to the fact they can differentiate them from real food. As well as this, gut-retention in copepods has been reported as just a few hours (Cole et al., 2013), with one study finding microplastics ingested by C. *finmarchicus* were egested within 2-4 hours (Vroom et al., 2017), so even if microplastics were ingested by these individuals, they would have been quickly egested. It is more likely that the faecal pellets from these individuals would act as a transport vector for microplastics to the deep ocean. In a study by Gunaalan et al (2023), faecal pellets from zooplankton were individually analysed and  $3 \times 10^{-3}$  microplastic per faecal pellet was observed.

Despite the possibility that copepods may be able to differentiate between microplastics and their prey, just over half of the stations sampled for this study contained individuals that had microplastics present. It may be that these microplastics were attached to food sources or simply ingested by mistake. A recent study reported particle concentrations of 1.4 to  $4.5 \times 10^3$  m<sup>-3</sup> in sea water samples taken in the Fram Strait (Bergmann et al., 2023), proving microplastics are readily available to marine biota. It has also been suggested that microplastics may overlap in size with phytoplankton, however as previously discussed, *C. finmarchicus* and other zooplankton are efficient at distinguishing between microplastics and prey (Xu et al., 2022, Gunaalan et al., 2023).

4.4.1.2 Seasonal variations in microplastic concentrations. Results from this study showed that in May, only samples from the surface contained microplastics (Fig. 3). We would expect to see a higher microplastic content in *C. finmarchicus* in May surface water samples, as this is the time of year when they ascend back to the surface after over-wintering to feed and reproduce (Jónasdóttir et al., 2015). No microplastics were found in individuals from deepwater samples in May, despite the same sample size. This is likely because these individuals are still in diapause, and any microplastics retained from feeding in the previous May will have been egested (Vroom et al., 2017).

There was a higher incidence of microplastics in individuals sampled during August, with both surface and deep-water samples containing microplastics. Most *C. finmarchicus* during this month will either still be feeding at the surface or will most likely have descended to deeper waters to enter diapause after accumulating large lipid stores (Jónasdóttir et al., 2015). It is therefore unsurprising to see a presence of microplastics in individuals sampled at the surface during this month, as they are still feeding. It is unlikely however, that we would see microplastics in *C. finmarchicus* once they have entered diapause. During this time, there is a pause in development and feeding, and a reduction in gut tissue and metabolic activity (Häfker et al., 2018). Any microplastics ingested during the previous spring bloom would likely be egested (Cole et al., 2013, Vroom et al., 2017). It is therefore interesting that microplastics were present in deep samples and shows that some individuals are retaining microplastics for longer periods of time. A likely explanation for this is that the microplastics were retained within the guts of the *Calanus* for longer or may have crossed the wall of the gut into the tissue. Due to the irregular sizing of microplastics in the environment, it is possible that these

particles can become entangled in the digestive system which may lead to greater retention times (Cole et al., 2013). There is also evidence to suggest microplastics can form aggregates in the gut, which could also lead to greater retention times (Vroom et al., 2017). It is difficult to determine the location of microplastics found in this study due to the entire animal being digested for analysis, but other studies have highlighted the potential for microplastics to remain in the gut or translocate to other tissues (Vroom et al., 2017), and it has been observed in other organisms such as mussels, where particles translocated from the gut to the circulatory system within 3 days and persisted for over 48 days (Browne et al., 2008).

It is unclear, however, why we are seeing microplastics present in individuals taken from deep water samples during August, but not in individuals taken from deep water samples during May. It is important to note the small sample size in this study, and the possibility that this may affect the reliability of the data shown. Sample sizes were dictated by the availability of *C. finmarchicus* at stage C5 within the water samples collected.

4.4.2 Microplastic polymer types and size. It has been found that aged microplastics are ingested at higher rates than pristine microplastics by C. *finmarchicus*, which may be attributed to the formation of a biofilm, mimicking prey (Vroom et al., 2017). It is possible that the polymers ingested by *C. finmarchicus* in this study had been in the environment for some time, most likely coming from long-range sources, transported through the Fram Strait via the West Spitsbergen Current.

Ethylene-vinyl-acetate (EVA) made up 67 % of the total particles found in *C. finmarchicus*, with polypropylene (PP) making up 22 % followed by chlorinated polyethylene (PE-Cl) at 11 % (Fig. 4). EVA has been found in multiple other studies in the Fram Straight (Peeken et al., 2018, Tekman et al., 2020). Near-surface samples located in the East Greenland Current region contained the highest proportion of EVA, with modelled trajectories indicating polar waters as the only likely origin. In this study, EVA particles were only present in surface samples suggesting, in line with previous studies, that this polymer is abundant in surface waters around Greenland and the Fram Strait. EVA is a widely used plastic, known for its flexibility, low-temperature toughness, and resistance to UV radiation. It has a density of 0.93–0.95 g cm<sup>-3</sup> and is mostly used in packaging for food and medical products, and in footwear, for example the soles of sports shoes and flip-flops (Material Properties, 2023). These main uses

suggest that EVA particles are likely to leach into the marine environment via direct disposal of packaging or weathering of shoes.

Chlorinated polyethylene (PE-CI) was only present in individuals from deep water samples. In a study investigating microplastic abundance in sediment samples from the Fram Strait, chlorinated polyethylene made up the largest proportion of microplastics (Bergmann et al., 2017), suggesting PE-CI is abundant in deep water and sediments in this area. Polypropylene is one of the most abundantly manufactured polymers (Erni-Cassola et al., 2019) so it is unsurprising that this polymer was also present. However, despite this being a low-density polymer, it was only present in individuals from deep water samples. Blank samples contained only one type of microplastic: artificially modified cellulose. The lack of polymer diversity in the blank samples suggests other polymers found in environmental samples did not come from contamination and are a true representation of polymer types in Arctic samples.

When discussing microplastic size, we must first consider the methods used for particle detection. A range of sizes are reported for microplastic pollution in the literature. Several studies have reported higher concentrations of microplastics in the smaller size ranges (e.g., Bergmann et al., 2017, Bergmann et al., 2019, Peeken et al., 2018). However, certain sampling techniques may exclude these smaller size ranges, especially when looking at microplastic particles <100 µm, which may be leading to an underestimation of the abundance of microplastics in this region. The identification methods used in this study had the capability of finding microplastic particles down to 25 µm, however unlike many studies looking at microplastic pollution, we found no particles <50 µm in both deep and surface samples (Fig. 5). This may have been due to the methods used for digestion, as the digested samples were filtered onto stainless steel mesh filters with a 50 µm pore size, so any particles less than this will likely have been lost. The highest concentrations reported for Arctic Sea ice thus far were found in the Fram Strait, and contained microplastics that were mostly <50 µm, with 67 % of these being in the currently smallest detectable size class of 11 µm (Peeken et al., 2018). However, the most common size class in water samples from the Fram Strait in a recent study was 100–200 μm, followed by <50 μm (Botterell et al., 2022). In this study, the majority of microplastics found were in the larger size ranges (101–150 μm and >150 μm), with the largest particle measuring 3.7 mm. Due to the size of *C. finmarchicus* (typically between 2 and 4 mm in length) we would expect to see fewer large particles ingested. It is possible that these larger

particles were fibres, which could have been coiled up and then extended during processing, although shape was not specifically assessed during this analysis.

#### 4.5 Conclusion

The investigation into the concentration of microplastics in the copepod species *C. finmarchicus* presents interesting results. Through sampling during two research cruises in May 2018 and August 2019, notable differences in microplastic abundance within *C. finmarchicus* populations from surface and deep waters emerged. Individuals in samples from surface waters contained microplastics exclusively during the May cruise, while microplastics were detected in individuals from both surface and deep waters in August. Results from this study suggest that the body burden of microplastics in *C. finmarchicus* is very low, and it is unlikely that they represent a significant transport vector for microplastics from surface waters to the deep ocean. However, the results do suggest that despite the short gut-retention time for *C. finmarchicus*, it is possible for microplastics to be retained for a longer period of time. The theories presented in this study are based on data for a single species and therefore present a conservative estimate of the potential importance of the role of zooplankton in transporting microplastic in the Arctic. Further research is needed to better understand seasonal dynamics and ecological implications.

# **Acknowledgements**

The samples in chapter 4 were collected onboard two research cruises on the RRS James Clark Ross: May 2018 (JR17005) and August 2019 (JR18007) by Daniel Mayor. I would like to thank Dan for his support with this chapter, and Kathryn Cook, Eloise Savineau, Katsia Pabortsava and Victoria Fulfer for their support during the laboratory analysis of these samples.

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# **Supporting Information**

Table S1: Sampling information including the month/year sampled, the depth (m) at which samples were taken, and coordinates for each sampling station, as well as microplastic count for each sample, with polymer type, blank corrected figures and the final microplastic number concentration per individual.

Samples were adjusted to compensate for the contamination of artificially modified cellulase by subtracting the average number of artificially modified cellulose particles found in the blank samples from samples containing the same polymer. The average number of artificially modified cellulose particles was 1.

Sampling station	Month / Year	Depth (m)	Latitude	Longitud e	MP count	Polymer type	Blank correcte d MP count	Final MP ind-1
1	May-18	974- 875	76.1156 6	-8.55815	0		0	0.00
1	May-18	125-5	76.1281 3	-8.72297	3	ethylene- vinyl-acetate	3	0.20
2	May-18	1000- 875	78.9945 1	-4.859	0		0	0.00
2	May-18	125-5	78.9852 8	-4.66726	1	ethylene- vinyl-acetate	1	0.07
3	May-18	1000- 875	78.9970 8	-2.4552	0		0	0.00
3	May-18	125-5	78.9917 5	-2.7258	0		0	0.00
4	May-18	1000- 875	80.3116 9	2.22461	1	Cellulose artificial	0	0.00
4	May-18	125-5	80.3399 6	2.4349	0		0	0.00
5	May-18	1000- 875	79.0016 9	1.83705	1	Cellulose artificial	0	0.00
5	May-18	125-5	79.0089 9	1.67294	0		0	0.00
6	Aug-19	875- 750	75.8304 1	-7.30614	1	Polypropylen e	1	0.07
6	Aug-19	125-5	75.8709 6	-7.4659	1	ethylene- vinyl-acetate	1	0.07
7	Aug-19	1000- 875	76.0341 8	-8.15439	0		0	0.00
7	Aug-19	125-5	76.0333 6	-8.16336	1	ethylene- vinyl-acetate	1	0.07
8	Aug-19	1000- 875	79.1630 9	6.8456	0		0	0.00

8	Aug-19	125-5	79.1598 5	7.17363	0		0	0.00
9	Aug-19	1000- 875	79.2785	2.60359	2	Cellulose artificial, Polypropylen e	1	0.07
9	Aug-19	125-5	79.2297 8	2.5351	0		0	0.00
10	Aug-19	1000- 875	78.3805 8	6.88534	1	Polyethylene, chlorinated	1	0.07
10	Aug-19	125-5	78.3140 3	6.76789	1	Cellulose artificial	0	0.00
11 (ice station)	Aug-19	0-200	78.9032 8	-0.30872	1	ethylene- vinyl-acetate	1	0.07
12 (ice station)	Aug-19	0-200	78.349	-4.6838	0		0	0.00

Table S2: Microplastic values for procedural blanks (n = 4).

Sample	MP count	Polymer type
Blank 1	0	
Blank 2	1	Cellulose
		artificial
Blank 3	2	Cellulose
		artificial
Blank 4	1	Cellulose
		artificial

# 5. Conclusions and areas of further research

The overarching aim of this research was to expand current knowledge on the occurrence, transfer, and fate of microplastics in the Arctic. Each chapter presents its own discussion, but this chapter aims to pull together the findings of the thesis and identify areas of potential future research. The results from this research uphold the views discussed in Chapter 1, that microplastics are ubiquitous in the Arctic, present in a range of environmental matrices and are subject to physical and biological processes that influence their distribution, concentration and potential impacts. Although one chapter was dedicated to examining microplastic occurrence in Arctic copepod, the concentrations were extremely low which raises questions regarding interactions between biota and abiotic matrices. For example, the transfer and timing of transfer of microplastics to the base of the marine food web, coupled to the seasonal life cycle of key organisms like copepods is not understood at all. Overall, these findings indicate that ongoing environmental change in the Arctic, particularly shifts in sea ice extent and seasonality, may alter both the transport pathways and ecological exposure of microplastics, underscoring the need for continued investigation into their long-term behaviour and impacts in polar marine systems.

5.1 Summary of results. The Arctic is a dynamic and complex environment, where pollutants are cycled throughout compartments such as snow, sea ice, seawater, and biota. Therefore, it is important when studying microplastic pollution in the Arctic to consider all of these interconnecting compartments. This thesis attempted to do this by considering both biotic and abiotic components.

Listed below are key findings from this thesis which build upon current research and introduce new evidence:

1. Quality control measures are essential when researching microplastics. There is no universally accepted method to account for field or laboratory-based contamination in environmental samples used for microplastic analysis, therefore contamination mitigation and quality controls vary widely between studies. One of the main concerns in the findings from Chapter 2 is the size of the plankton mesh versus the sizes of the microplastics that were collected. Although it is plausible that smaller microplastics

could have become trapped on the mesh by the presence of other material e.g. organic particulate matter (like phytoplankton), it is likely that the particle numbers are biased to the larger size fractions as smaller particles (e.g. <100  $\mu m$ ) will not have been as efficiently retained during the filtration process. Rigorous methods were put in place to mitigate contamination in both the collection and processing of the seawater samples, and blanks were taken to account for contamination and microplastic numbers were corrected accordingly. Thus, we must assume that the observed microplastics concentrations represent real environmental concentrations. However, polypropylene (one of the most common polymers in global production) was evident in both the blanks and environmental samples and caution is urged when interpreting the environmental signal for this polymer given its ubiquitous occurrence in the blanks.

2. Microplastics are present throughout the water column. Microplastics were found in both surface and subsurface waters of the Beaufort Sea, a region with very few previous observations of microplastic pollution, despite its location making it a likely accumulation zone. There was no significant difference in microplastic number concentration and depth, although subsurface water samples did contain slightly more particles, suggesting that microplastics in these layers of the water column originated from the same source, or that microplastics are well mixed within this water column. The dominant plastic polymer found in samples was polypropylene, which is one of the most common polymers in production and has been found in other locations across the Arctic Ocean and coastal seas. There were no significant differences in microplastics abundance or polymer composition between the different sampling depths, again suggesting microplastics originated from the same source as different water masses (surface, polar mixed layer versus deeper water) did not show differences in particle type or number. The high numbers of microplastics found in this study indicate that sea ice in this region is also likely to contain high microplastic number concentrations, based on limited observations in Arctic sea ice but also the efficient transfer of microplastic particles into forming sea ice based on the sea ice chamber experiments conducted in this thesis. There is still a lack of knowledge on the behaviour and fate of microplastics in the Arctic Sea ice system.

- 3. Forming sea ice serves as a temporary sink for microplastics particles present in **surface seawater.** Results from chapter 3 show that particles become entrained in ice resulting in a spike of microplastics pollution in the upper layers of the ice with a corresponding significant decrease of microplastics in seawater when ice is present. Upon melting, the microplastics are released back into the surface water. Upon release however, there is evidence that microplastic particles accumulate in a lowersalinity water layer directly below the ice during melt, with implications for enhanced exposure for organisms associated with the underside of the ice. However, all of the polyethylene particles were perfectly or near-perfectly spherical. Results from studies investigating microplastic pollution in the Arctic show a dominance of fibres, followed by fragments. Particle morphology is likely to be an important factor in controlling microplastic transfer and accumulation in ice and the results from chapter 3, using only engineered microplastics spheres, should hence be used with caution. Nonetheless, the range of densities and sizes used at least provide a better understanding of the mechanistic processes driving the transfer of microplastics between seawater and sea ice. Furthermore, the study highlights that microplastics can become enriched in surface ice layers, possibly explaining the very high number concentrations reported in several Arctic sea ice studies. During ice melt, the observation of high numbers of microplastic particles in a meltwater layer beneath the ice pack raises the possibility of seasonally induced exposure for sympagic organisms.
- 4. Microplastics are present in Arctic Copepods at low concentrations. Microplastics concentrations in *C. finmarchicus* samples from the Fram Strait were very low, suggesting they are unlikely to be acting as a transport mechanism for microplastics as they move from feeding at surface waters during the spring bloom, to deeper water as they descend into diapause. Individuals in surface water samples from both the May 2018 and August 2019 cruises contained microplastic, whereas only individuals in deep water samples from the August 2019 cruise contained microplastics. However, observations of the differences in microplastic incidence between depths were not significant. There was no observed difference either between the months of sampling. The findings in this study indicate that microplastic pollution is very low for this species of zooplankton. However, it is important to note that this study was limited to a small

- sample number of Calanus in a distinct geographical region which may have impacted the ability to observe consistent patterns.
- 5. Variations in sampling and analytical methodologies impedes the comparison of results from other studies. Microplastic research has developed rapidly over the last decade, with more recent studies examining the effects that microplastics are having on marine organisms with possible effects on ecosystem functioning. However, due to this rapid development, there is a lack of uniformity in the methodology used for sampling and quantifying microplastics. In a study by D'Angelo et al (2023), similar sampling methods to those used in this study were employed, and results for microplastic concentrations in the Beaufort Sea were comparable. However, one marked difference was that the D'Angelo study quantified microplastics from the filtered water samples, whereas this study processed the actual filters which required the extra step of sample digestion. Also, the D'Angelo study did not conduct blank assessments to mitigate atmospheric (and other) contamination during sampling, and consequently fibres and all other microplastic categories were considered separately. The average concentration for plastic particles across all sampling stations, excluding fibres, ranged from 1.9 L<sup>-1</sup> to 11.7 L<sup>-1</sup>. The average concentration of plastic particles for the Beaufort Sea was  $1 \pm 1$  L<sup>-1</sup>. The slightly lower concentrations in the D'Angelo et al (2023) study compared to this thesis may be explained by the exclusion of fibres. In a study by Ross et al (2021), much lower concentrations of microplastics were reported for the Beaufort Sea despite sampling much higher volumes of water. Similar to this study, CTD/Rosette sampling was used to collect water column profile samples. However, sample identification involved a two-step analysis comprising of visual identification of suspected microplastics followed by a single point μFT-IR analysis to confirm that the suspected microplastics were indeed microplastics, rather than spectral analysis of the entire sample which was conducted for this research. It is also important to highlight the size detection limit, as this is often dictated by sampling methods or identification techniques. Therefore, caution must be taken when directly comparing results to other studies, and future research should consider reporting a particle size range along with total particle concentrations

This research attempts to create a holistic view of microplastics in the Arctic by providing microplastic quantities in biotic and abiotic components and a good illustration of the range of studies (using different sampling and analytical methodologies) is presented in Chapter 1 (review of Arctic observations). There are still many unanswered questions when looking at microplastic pollution in the Arctic. However, sampling associated with fieldwork in the Arctic is not easy and often biased to summer, ice-free periods of the year. The research conducted for this thesis benefited from samples collected during various successful field campaigns in the Arctic by other researchers. This was achieved through partnerships with individuals and institutions, highlighting the importance of cross-cutting work with other scientists and including Knowledge from other disciplines such as oceanography, marine biology, cryospheric science and atmospheric sciences will be key to understanding microplastic fate in the Arctic.

- 5.2 Areas for further research. The work presented in this thesis advances our understanding of the transport and fate of microplastic pollution in the Arctic, but there are still substantial knowledge gaps within this area of research and further work is needed to tackle these.
  - Studies on the effects of microplastic ingestion on Arctic biota, particularly for iceassociated (sympagic) biota. It is plausible that seasonal melt of sea ice creates a
    meltwater buoyant layer directly beneath ice floes that is enriched in microplastic
    particles thus providing a significant exposure route to organisms present in this
    environment. Furthermore, ice algae present on the underside of ice particularly in
    late winter following Polar Sunrise, may accumulate plastic particles released through
    gravity brine drainage and/or percolating meltwater. Both field and laboratory-based
    studies are needed to assess the accumulation and transfer of microplastics to the
    base of the pelagic food web and investigate the timing and magnitude of particle
    release from ice in relation to biological phenomena such as phytoplankton blooms.
  - A better assessment of potential 'sinks' for microplastics pollution e.g. sea ice, deep-sea sediments, ocean gyres, as well as a better understanding of microplastic sources to the Beaufort Sea, particularly deposition from the atmosphere and the role of the Mackenzie River in contributing plastics to surface waters is now required. This will require environmental monitoring in combination with the derivation of fluxes or

- loads from these sources. Future studies should aim to use cohesive sampling and processing methods to ensure results can be directly compared.
- The results displayed in this thesis demonstrate the advantages of using laboratory facilities such as mesocosms and chamber studies as a proxy to simulate Arctic environments and processes. In laboratory studies, such as chapter 3 of this thesis, environmental conditions and variables can be closely controlled. There are an array of experiments that could be conducted in the sea ice facility to further examine the mechanisms of microplastic fate in the sea ice system. For example, the use of plastic fragments and fibres instead of engineered spheres would provide a more realistic simulation of the Arctic marine system given observations of these plastic morphologies in the Arctic. Their uptake and fate in ice may be very different to the microspheres used in Chapter 3. Furthermore, the chamber system used in chapter 3 was essentially 'clean' with no organic matter (dissolved or particulate matter) or biota. Future studies could incorporate these components for a more realistic simulation of Arctic surface waters.
- The role of the atmosphere in contributing micro and nanoplastics to Arctic marine surfaces is not well understood, particularly in relation to observations of microplastics in Arctic seawater. The observations to date in snow samples have demonstrated the prevalence of tyre-wear particles (TWP) (e.g. natural rubber and styrene butadiene rubber) over other plastic polymers and the fate of TWP in the Arctic marine system is poorly understood. Atmospheric transport to the Arctic and subsequent deposition of other polymer types (e.g. PE, PP and PS) requires more detailed observations through systematic monitoring in different geographic locations, perhaps in conjunction with the existing Global Atmospheric Watch sites in the Arctic.