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Published in: Environmental Pollution

Link to article, DOI: 10.1016/j.envpol.2020.115248

Publication date: 2020

Document Version Peer reviewed version

Link back to DTU Orbit

Citation (APA):

Rist, S., Vianello, A., Winding, M. H. S., Nielsen, T. G., Almeda, R., Torres, R. R., & Vollertsen, J. (2020). Quantification of plankton-sized microplastics in a productive coastal Arctic marine ecosystem. *Environmental Pollution*, 266, Article 115248. https://doi.org/10.1016/j.envpol.2020.115248

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1 Quantification of plankton-sized microplastics in a

² productive coastal Arctic marine ecosystem

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14 Abstract

Microplastics (MPs) are polluting the Arctic, but our understanding of their abundance, distribution, 15 and sources is limited. This study quantified MPs down to 10 µm in marine waters of the most 16 populated region in Greenland. A new plastic-free pump-filter system was used to collect MPs from 17 surface waters in the ford Nuup Kangerlua close to Nuuk. Additionally, we took samples by 18 horizontal tows with a bongo net (300 µm mesh-size). The median concentrations were 142 MPs m⁻ 19 ³ and 0.12 MPs m⁻³ in the pump and bongo samples, respectively. The most abundant polymer was 20 polyester across stations and sampling types. Fibers were the dominant shape in the bongo samples, 21 while non-fibrous particles dominated in the pump samples. MP abundance was lower in the fjord 22 and increased close to Nuuk and towards the open ocean, indicating that Nuuk is an important point 23 24 source for MPs. In both samples, concentrations of MPs increased with decreasing size, illustrating the importance of quantifying the smallest fraction of MPs. Thus, the use of methods allowing for a 25 quantification of the smallest MPs is vital to reduce the underestimation of MP concentrations in the 26 27 environment. The smallest size fraction is also most available to plankton-feeding marine invertebrates and an important entry point for MPs into marine food webs. At the found 28 29 concentrations, immediate adverse effects on the pelagic food webs are unlikely. However, growing anthropogenic activities could increase the risk of MPs to affect the sensitive Arctic ecosystem. 30

31

32 Capsule:

Sampling of microplastics >10 µm in West Greenland with a new pump system demonstrated the
dominance of the smallest size fraction, which is the most likely to enter pelagic marine food webs.
Keywords: Greenland; plastic pollution; pump-filter system; bongo net; µFTIR-Imaging

36 Introduction

It is well documented that microplastics (MPs, <5 mm) have already polluted the most remote areas 37 on earth, such as the polar regions (Cincinelli et al., 2017; Lusher et al., 2015; Obbard et al., 2014). 38 In the Arctic, MPs have already been detected in marine waters (Morgana et al., 2018; Tekman et 39 al., 2020), sediments (Bergmann et al., 2017), sea ice (Peeken et al., 2018), and snow (Bergmann et 40 al., 2019). Although data on MP concentrations in the Arctic is still very limited, a wide range of 41 concentrations has been reported for Arctic seawater: from 0.0007 to 31,300 MPs m⁻³ (Table 1). 42 Especially in sea ice and snow surprisingly high concentrations have been documented; for instance 43 up to 1.2·10⁷ MPs m⁻³ in ice cores of Fram Strait (Peeken et al., 2018). The total load of plastics 44 45 floating on the surface of the Arctic Ocean has been estimated to range between 100 and 1200 tons 46 (Cózar et al., 2017), and global modeling of the distribution and accumulation of marine debris identified a garbage patch in the Barents Sea (van Sebille et al., 2012). These high levels of plastic 47 48 pollution in an area with extremely low direct anthropogenic impact can to a great extend be explained by long-range transport of plastics with air and currents (Obbard, 2018). Atmospheric 49 deposition has only recently been identified as a major pathway for global MP transport (Brahney et 50 51 al., 2020) and can most likely explain high concentrations of MPs in Arctic snow samples (Bergmann et al., 2019). In contrast, long-distance transport by currents is well known and the 52 poleward branch of the Thermohaline Circulation is a major pathway of plastics from the North 53 Atlantic to the Greenland and Barents Sea (Cózar et al., 2017). Model simulations suggest that these 54 areas are accumulation zones for plastics, holding 95% of the estimated plastic load in the Arctic. 55 This was corroborated by field measurements (Cózar et al., 2017). One branch of the Thermohaline 56 Circulation also goes into the Labrador Sea, but except for the study by Cózar et al. (2017), no data 57 on MP pollution is available from the west coast of Greenland. 58

59 West Greenland receives plastic debris from the North Atlantic as well as from local point sources. Relevant point sources of plastic pollution include fishing and maritime industry, raw sewage 60 outlets, mismanaged waste, and last but not least Nuuk, the capital and largest city of Greenland 61 with 18,000 inhabitants. Nuuk holds the biggest port of Greenland and the city's wastewater is 62 63 discharged untreated to the fjord Nuup Kangerlua (Gunnarsdóttir et al., 2013). Thus, there are several unquantified sources of MP pollution in West Greenland. If MPs introduced to the marine 64 ecosystem are taken up by the marine food web, Greenlandic livelihood may be impacted. Fishery 65 alone provides more than half of Greenland's export income (Jacobsen et al., 2018). If the food web 66 efficiency or the quality of the marine products are affected by MPs, it will potentially have a major 67 impact on the local and national economy of Greenland. 68

69 The present study is the first to focus on marine MPs in the most populated area of Greenland. Furthermore, we applied a new plastic-free pump-filter system, which allows the collection and full 70 71 quantification of MPs down to 10 µm. Very few studies have quantified MPs down to that size in the Arctic (Table 1). This abundant and understudied fraction of MPs overlaps in size with the 72 planktonic prey of the dominant secondary producers in the Arctic, the Calanus copepods (Cole et 73 74 al., 2013; Levinsen and Nielsen, 2002). The large lipid-rich Calanus copepods have a key position in the Arctic marine ecosystem. They provide food for the important Greenlandic fish stock and are 75 responsible for carbon sequestering through the export of carbon to the deep waters and sediment 76 77 by the production of large, fast-sinking fecal pellets (Juul-Pedersen et al., 2006). The presence of plankton-sized marine MPs in the highly productive coastal ecosystem around Nuuk gives rise to 78 concern for the quality of the important commercial and recreationally exploited fish stocks 79 80 (Jacobsen et al., 2018). Therefore, the present study aims to provide a baseline of plankton-sized MPs (>10 µm) from the fjords of Greenland and the coastal area towards the potentially most 81 82 important point source of MPs in West Greenland: the capital Nuuk.

84 Materials and Methods

85 Study site

The sampling of six stations in Nuup Kangerlua, in the bay in front of Nuuk, and at the banks off 86 the coast of West Greenland, was done from the research vessel R/V Sanna (Greenland Institute of 87 Natural Resources) between 10 and 12 of May 2019. The stations differed in their proximity to the 88 89 expected MP point source Nuuk (Fig. 1, see more details in SI). Nuup Kangerlua is located on the 90 southwest coast of Greenland with the capital Nuuk (64°10'N, 51°44'W) located at the mouth of the fjord. The average depth is approximately 250 m (max. depth >600 m) (Mortensen et al., 2011). 91 92 The main fjord branch is approximately 190 km long, and several sills characterize the outer part of 93 the fjord. The uppermost branch of Nuup Kangerlua connects the marine environment to the margin 94 of the Greenland Ice Sheet. The freshwater input induces a seasonal stratification of the upper part of the water column (Fig. 1), which is further strengthened by solar heating during summer 95 96 (Mortensen et al., 2013, 2011).

97 CTD measurements

On arrival at the sampling station, vertical profiles of water temperature, salinity, density, and 98 99 fluorescence were obtained using a CTD (SBE 19plus V2 CTD from Sea-Bird Electronics) 100 equipped with a Seapoint chlorophyll a fluorometer and a Biospherical/Licor sensor. Depth profiles were recorded from the surface to 5 m above the bottom. Water samples for chlorophyll a (chl a) 101 measurements were collected at 1, 5, 10, 15, 20, 30, 40, 50, 100, 150, 250, and 300 m depth using a 102 5 L Niskin bottle between 1 and 2 of May. Each sample was filtered onto a GF/F filter and extracted 103 in 96% ethanol for 12-24 h (Jespersen and Christoffersen, 1987). Fluorescence was measured on a 104 105 Turner TD-700 fluorometer (Turner Designs, California, USA) before and after acidification. The

fluorometer was calibrated against a pure chl *a* standard. The analysis was used for calibration of the CTD fluorometer, and chl *a* concentrations were then calculated from CTD fluorescence profiles at all stations. The phytoplankton biomass was calculated from the average chl *a* in the upper 10 m, a carbon/chl *a* conversion factor of 42.6 (Juul-Pedersen et al., 2006) and a carbon to dry weight conversion factor of 55% (Hansen et al., 1994).

111 MP sampling

Two different sampling devices were employed to collect one sample each at the six stations. We 112 collected MPs >10 µm from 5 m depth using a custom made pump-fed filtering device (hereinafter 113 termed "pump samples"). In addition, we took surface samples with a submerged bongo net 114 targeting >300 µm sized MPs (hereinafter termed "bongo samples"). The plastic-free pump-filter 115 116 system was developed to sample the smallest fraction of MPs, which is often neglected in other studies. The purpose of the bongo samples was to allow comparability to previous studies using this 117 118 and similar net-sampling techniques. Both samples were taken in the upper 5 m, i.e., above the pycnocline (Fig. 1), in the water impacted by the meltwater from the inner part of the fjord system. 119 Unfortunately, the bongo sample from station 1 was lost during transportation, resulting in only 5 120 121 stations for this sampling method.





The plastic-free pump-filter device (UFO system - Universal Filtering Objects system) was 125 composed of a metal hose deployed in the water, a pump controlled by an inverter, and a modular 126 filtering device capable of filtering large volumes of water (Fig. S1, SI). All parts of the system 127 were in metal but for a gasket of rubber in the three filtering cartridges themselves. In brief, the 128 water was pumped through a flexible metal hose, the mouth of which was equipped with a stainless 129 steel metal cage of 5 mm mesh to protect the system against large debris. A positive displacement 130 pump with a brass impeller (Oberdorfer Gearpump N994RE) sent the water through a short metal 131 hose to the filter cartridges. Three separate stainless steel filter cartridges were applied, which each 132 contained a stainless steel filter of 167 mm diameter. The water first passed through a filter of 300 133 134 µm mesh to retain bigger items with the purpose of protecting the finer filtering mesh from clogging. The water was then divided onto two parallel units with filters of 10 µm. The outlets were 135

136 re-combined and connected to a mechanical flowmeter to quantify the filtered volume. The inlet hose was deployed at 5 m depth using a crane, and approximately 1 m³ of water was filtered at each 137 station. After sampling, the system was evacuated, and the single filter cartridges were opened 138 inside a clean fume hood to prevent plastic contamination. Both the 300 µm and the two 10 µm 139 enriched filters collected at each station were transferred to a glass petri dish, which subsequently 140 was wrapped in aluminum foil and frozen at -20°C until sample processing. Three air blanks were 141 taken during the sampling by exposing empty petri dishes with pre-muffled 10 µm steel filters to 142 the surrounding environment during the opening phase of the UFO system. The air-blanks were 143 taken and stored similarly to the samples. Moreover, several paint fragments were collected 144 onboard the ship to assess the potential contamination related to this specific source. 145

The bongo net, equipped with a flowmeter, was deployed 3 m from the side of the ship using a crane and dragged for 20 min in the upper 0.5 m of the water column. The net was made of nylon, it had a mesh size of 300 μ m, and the samples were collected in the cod-end of the net. The bottle at the cod-end was removed from the net, and the content was carefully rinsed onto a 300 μ m nylon mesh. The mesh was carefully folded, wrapped in aluminum foil, and stored in a polyethylene plastic zip-lock bag at -80°C until sample processing.

152 Sample processing

The pump samples were processed following a protocol slightly modified from Liu et al. (2019)
(see SI for details). In brief, this included six treatment steps: 1) SDS (5% sodium dodecyl sulfate)
treatment for 24 h and transfer of samples onto 10 µm steel filters (47 mm diameter), 2) overnight
incubation with protease (Sigma, Protease from *Bacillus sp.*) and lipase (Strem Chemicals,
Lipozyme® TL 100L), 3) incubation with cellulase (Sigma, Cellulase enzyme blend) and
Viscozyme® L (Sigma) for 72 h, 4) oxidation (Fenton reaction) with hydrogen peroxide, sodium

159 hydroxide and iron sulfate overnight, 5) size fractionation with a 500 µm steel sieve, 6) density separation of the fraction $<500 \,\mu\text{m}$ with sodium polytungstate (1.9 g cm⁻³). The fraction $>500 \,\mu\text{m}$ 160 underwent spectroscopic analysis together with the bongo samples. The filter with the sample 161 fraction <500 µm was sonicated and rinsed with 50% ethanol, and all liquid was sequentially 162 163 transferred to 10 mL glass vials and evaporated in a water bath at 50°C using a stream of nitrogen (Biotage, TurboVap). When the samples were dry, 5 mL of 50% ethanol were added for subsequent 164 analysis with FPA-µFTIR-Imaging (Focal Plane Array-Fourier-Transform Infrared Imaging-micro 165 Spectroscopy). The collected air-blanks from the ship underwent the same processing and analysis 166 in the laboratory as the pump samples. Thus, the blank samples monitor the combined MP 167 contamination potentially occurring onboard and later during sample processing and analysis. 168 169 For the bongo samples we used a simplified protocol. The samples were rinsed off the mesh into glass beakers with a 5% SDS solution and kept at 50°C with gentle stirring (100 rpm) for 48 h. The 170 171 samples were then sieved over a 200 µm stainless steel sieve, rinsed with filtered water, and flushed back into the beaker with 200 mL of Tris (trisaminomethane)-HCl buffer (pH 8.3), adding 700 µL 172 of protease, 500 µL of lipase, and incubating at 50°C with gentle stirring for 72 h. Thereafter, 173 samples were vacuum-filtered onto 10 µm stainless steel filters (47 mm diameter) and stored in 174 glass petri dishes until analysis. 175

176 MP analysis

The pump samples were analyzed using FPA-μFTIR-Imaging, which is, at present, considered the
most suitable analytical approach for analysis of small MPs (Liu et al., 2019; Löder et al., 2015;
Mintenig et al., 2016; Primpke et al., 2016; Simon et al., 2018; Vianello et al., 2019). A sub-sample
of the 5 mL particle suspension (3 aliquots of 1-1.2 mL – corresponding to 60-72% of the total
volume) was deposited onto three 13×2 mm zinc selenide (ZnSe) infrared windows pre-heated and

182 held in a compression cell (PIKE Technologies, Fitchburg, WI, USA) using a capillary glass pipette (micro-classic, Brand GmbH, Germany). Each enriched window was dried overnight (55°C) and 183 subsequently analyzed, accounting for the whole active surface (10 mm diameter - 78.5 mm²). The 184 applied instrument was an Agilent 620 FTIR microscope with a 128×128-pixel FPA detector 185 186 combined with a Cary 670 FTIR spectrometer (Agilent Technologies, Santa Clara, CA, USA). It provides two main outputs: a magnified optical image, and the relative IR map of stitched tiles of 187 128×128 pixels, co-adding several scans. Each IR map pixel contains an FTIR spectrum, allowing 188 the identification of a wide range of organic and inorganic materials (including synthetic polymers) 189 by comparing the unknown spectra with a dedicated database. The analysis was carried out in 190 transmission mode in a spectral range of 3750-850 cm⁻¹ at 8 cm⁻¹ resolution, using a 15x Cassegrain 191 objective/condenser with 5.5 µm resulting pixel size; 120 co-added scans were collected for the 192 background on a single tile, while 30 co-added scans were recorded when scanning the sample. The 193 beam attenuation was 50%. The scan time at these settings was approximately four hours. 194 The bongo samples were visually inspected using a stereo microscope with a connected camera 195 (SteREO Discovery V8 with Axiocam 105 Color, Zeiss GmbH, Oberkochen, Germany). All 196 197 potential plastic particles were photographed and categorized by shape (fibers vs. all other shapes), length, and width. The material composition of each particle was then analyzed by Attenuated Total 198 Reflection - Fourier Transform Infrared Spectroscopy (ATR-FTIR), using an Agilent Cary 630 199 FTIR spectroscope (Agilent Technologies, Santa Clara, CA, USA) with a single reflection diamond 200 201 ATR. The samples were positioned onto the ATR crystal and then compressed using the instrument clamp to achieve optical contact, allowing to record surficial ATR-FTIR spectra. The spectra were 202 recorded at 4 cm⁻¹ spectral resolution by co-adding 64 scans, and a background in air was collected 203 204 before each measurement. Each collected spectrum was exported and compared to multiple spectral

databases containing both synthetic polymers and non-synthetic materials (Omnic 8, Thermo Fisher
Scientific, Madison, WI, USA).

207 Contamination control

The use of plastic materials during sampling and sample processing was avoided if possible, except 208 for the polytetrafluoroethylene (PTFE) stopcock of the separation funnels and the PTFE septa of the 209 vials containing the processed pump samples. Therefore, this material, together with the rubber 210 from the UFO gaskets, was excluded from the MP quantification. Furthermore, we took samples of 211 the ship's paint and excluded matching paint particles in the samples. For the bongo samples, 212 potential contamination could have resulted from the nylon mesh as well as the polyethylene zip-213 214 lock bags. All materials were either rinsed with filtered water or muffled at 500°C and wrapped in aluminum foil until use. Samples were also covered with aluminum foil or glass lids whenever 215 possible. All solutions used for processing the samples were filtered over 0.7 or 1.2 µm GF filters. 216 Sample processing was done in a laminar flow cabinet, and cotton lab coats were worn. As 217 described above, three air-blank samples were collected on the ship during sampling, which 218 underwent the same laboratory processing and analysis as the real samples. They therefore represent 219 the total contamination, from sampling until sample analysis. 220

221 Data analysis

The collected FPA-Imaging data were processed using siMPle (Primpke et al., 2020), an opensource software developed by Aalborg University and Alfred Wegener Institute, which allows the automated analysis of large µFTIR-Imaging datasets. In brief, it runs a Pearson correlation between each sample spectrum and the database using the raw spectrum, first and second derivative, reconstructing each particle from the FTIR spectra of the pixels it covers, and hence providing a false-color map of the identified materials in the sample. The software also provides morphological

and size measurements (Vianello et al., 2019), as well as estimated mass measurements (Liu et al.,
2019). With regards to particle shape, we differentiate between fibers and particles (i.e. all other
shapes) as described in Vianello et al. (2019) (Fig. S3, SI). The mass of the plastic particles in the
pump samples was estimated following an approach described by Simon et al. (2018). In short, the
2-dimensional size and shape of a MP particle obtained by the siMPle analysis are used to estimate
a particle volume. The mass is then derived by multiplying with the typical density of the
determined polymer.

235

236 **Results and Discussion**

237 MP abundance and size distribution

We found MPs in the water samples from all stations. In the pump samples (>10 µm), we found MP 238 concentrations of 67-278 particles m⁻³, with a median of 142 particles m⁻³ (Fig. 2a). The MP 239 concentrations (>300 µm) found in the bongo samples were 2-3 orders of magnitude lower than 240 those collected by the pump, ranging between 0.08 and 0.4 MPs m⁻³ with a median concentration of 241 0.12 MPs m⁻³ (Fig. 2b). Data on MPs in Arctic waters is scarce, and a wide range of concentrations 242 has been reported (Table 1). The results of the bongo samples are in a similar range as what was 243 collected using a Manta net around Svalbard (Lusher et al., 2015) and in the Chukchi and Bering 244 Sea (Mu et al., 2019) as well as from pump samples in the Arctic Central Basin (Kanhai et al., 245 2018). The MP concentrations from our pump samples are comparable to recently reported values 246 from surface water samples in the Fram Strait, where 113-262 particles m⁻³ were found (except for 247 one station with 1287 particles m⁻³) (Tekman et al., 2020). Tekman et al. (2020) fully quantified 248 MPs down to 32 µm (the size of the sieve during sampling) and semi-quantified MPs down to 11 249 250 μm.

Despite the comparative remoteness of the studied area, the concentrations of MPs found in this study were similar to those reported in locations with higher anthropogenic activity. In Manta trawls close to the Danish coast, the average MP concentration was 0.07 m^{-3} (>300 µm) (Tamminga et al., 2018), and in marine waters around Japan, Isobe et al. (2015) found a median concentration of 0.74 MPs m⁻³ (>350 µm). A quantification of MPs down to 10 µm in a transect from the European coast across the Atlantic to the Sargasso Sea gave 13 to 501 MPs m⁻³, with the highest concentrations in the English Channel (Enders et al., 2015).

Table 1. Reported MP concentrations in seawater, sea ice and snow samples in the Arctic region.

	Matrix and	MP conc.	Size limit	Sampling method	Size limit		
Location	depth	(MPs m ⁻³)	(µm)		Dominant polymer	Reference	
Nuup Kangerlua, West	Water, 5 m	142	10	Pump		This	
Greenland	Water, surface	0.12	300	Bongo net	Polyester	study	
Greenland Sea Gyre	Water, surface	800-3740	50	Pump and plankton net	Polyester	[1]	
East Greenland	Water, 0-50 m	2.38 (2014)	500	WP-2 net	Polyester	[2]	
Northeast Greenland	Water, 6 m	2.4	80	Water intake	Polyethylene	[3]	
Greenland & Barents	XX / C	(2.1041 -2	500			[4]	
Sea	Water, surface	6.3·10 ⁺ km ⁻²	500	Manta trawl	-	[4]	
Fram Strait	Water, 1-5350 m	95	32	McLane WTS-LV	Polyamide	[5]	
Fram Strait, Svalbard	Snow	$1.76 \cdot 10^{6}$	11	Sampled with a spoon	Acrylates/PUR/varnish	[6]	
<u> </u>	Water, surface	0.34	333	Manta trawl		[7]	
Svaldard	Water, 6 m	2.68	250	Onboard pump	Polyester, polyamide	[/]	
	Water, 0-1000 m	0.0007-0.048		Niskin bottles	Paint	[8]	
Svalbard*	Sea ice, free float	0.158	50	Boat hook	Unknown pigment		
	Water, 8.5 m	0.7	250	Water intake	Polyester	[9]	
Arctic central basin	Water, 8-4369 m	20.8		Niskin bottles			
Arctic	Water, surface	31,300	100	1 L grab samples	Polyester	[10]	
Central Arctic	Sea ice	0.38-2.34.105	0.22	Drilling of ice cores	Rayon	[11]	
Arctic	Sea ice	0.11-1.2.107	11	Drilling of ice cores	Polyethylene	[12]	
Arctic Central Basin	Sea ice	0.2-1.7·10 ⁴	100	Drilling of ice cores	Polyester	[13]	

	Water below ice	0-18	250	Pump and sieve		
Chukchi Sea	Water, surface	0.23	330	Manta trawl	Polyethylene	[14]
Bering Sea		0.091			terephthalate	

*Quantified all anthropogenic particles, including non-synthetic. [1] (Jiang et al., 2020), [2] (Amélineau et al., 2016), [3] (Morgana et al., 2018), [4] (Cózar et al., 2017), [5] (Tekman et al., 2020), [6] (Bergmann et al., 2019), [7] (Lusher et al., 2015), [8] (von Friesen et al., 2020), [9] (Kanhai et al., 2018), [10] (Barrows et al., 2018), [11] (Obbard et al., 2014), [12] (Peeken et al., 2018), [13] (Kanhai et al., 2020), [14] (Mu et al., 2019)

260

261 Other studies from the Arctic report both lower and higher MP concentrations in comparison to our findings (Table 1). Several factors, however, complicate comparisons between studies. Firstly, the 262 Arctic covers a vast area and the sampled locations differ greatly with regards to oceanographic 263 regimes and potential MP sources. Since our study is the first to analyze MPs on the west coast of 264 Greenland, the closest available data on MPs comes from East Greenland. There, long-range 265 266 transport is expected to play a much bigger role, while local sources are more relevant for West Greenland (see MP distribution and sources). Secondly, studies employed widely different 267 methodologies for sampling, sample preparation, and analysis, as well as the analyzed size 268 fractions. The problems with lack of standardization are frequently emphasized within the MP field 269 (e.g., Avio et al., 2017; Mai et al., 2018). The impact of the chosen methodology on the results 270 becomes very apparent when comparing our two sampling methods. The concentrations from the 271 272 pump and the bongo net differed by 2-3 orders of magnitude. This is likely related to several factors. While the particle identification and quantification of the pump samples were fully 273 274 automated, the analysis of the bongo samples relied on visual pre-sorting, which entails the risk of missing particles. More importantly though, the lower size limit was very different, with 10 µm for 275 the pump and 300 µm for the bongo net. The results of our pump samples clearly show an increase 276 in particle numbers with decreasing size (Fig. 2e). The size distribution was similar for all stations 277 (Fig. S2, SI). Overall, 93% of all the particles we found were smaller than 300 µm and 68% smaller 278 than 100 µm. Only for the size fraction of 11-25 µm we found very few particles. This is most 279 likely related to the detection limit of the µFTIR approach, which is limited to around 11 µm due to 280

diffraction phenomena. The spectra of very small and thin particles often have a low signal-to-noise 281 ratio, affecting their automatic identification by the siMPle software, probably leading to an 282 283 underestimation of the smallest MP particles. The size distribution in the bongo samples showed a similar trend of higher particle numbers with decreasing size, with the exception of a large number 284 of very big particles (>5000 µm) (Fig. 2f). These were mainly long fibers, which were also the 285 286 dominant particle shape in the bongo samples (see MP shape and polymer composition). Similar size distributions as in our pump samples were found in studies on Arctic sea ice (Peeken et al., 287 288 2018), snow (Bergmann et al., 2019), and water samples (Tekman et al., 2020), which also analyzed MPs down to 11 µm. This finding clearly illustrates the importance of quantifying the smallest 289 MPs. Since the majority of published studies on MP concentrations in marine waters used a mesh 290 291 size that is considerably larger, concentrations are generally underestimated at present. Therefore, 292 we encourage the use of pump-filter systems as used in the present study or similar devices, which enable sampling the smallest size fraction of MPs that can be analyzed. 293





Figure 2. Microplastics (MPs) from pump (left panels) and bongo (right panels) samples: MP concentration (MPs m⁻³) in (a) the pump and (b) the bongo samples; Polymer composition (%) in (c) the pump and (d) the bongo samples; Size distribution (length - μ m) in (e) the pump and (f) the bongo samples. The red line in (e) and (f) shows the relative cumulative frequency of MPs.

300 Blank samples

301 The blank samples contained both the on-site airborne contamination and the contribution derived from the laboratory. The results showed an average contamination of 6.7 ± 2.5 MPs per sample, 302 corresponding to $4.8 \pm 2.5\%$ of the average MP abundance in the analyzed samples. The polymeric 303 composition of the contaminating MPs was 50% polyester, 25% polyamide (nylon), 17% ethylene 304 305 vinyl acetate, and 8% polystyrene. The results were corrected for contamination by subtracting the averaged contribution of every single polymer found in the blank samples (both by particle number 306 307 and mass). The polyester and nylon items identified in the blank samples could be related to 308 airborne contamination from textiles, as most of the technical clothes used onboard are made of these polymers. The ship paints were another relevant source of contamination found in the 309 analyzed samples, but not in the blanks. Several paint particles were identified by µFTIR-Imaging 310 analysis as the specific paints collected onboard from different surfaces of the ship during the 311 survey. This specific contribution was excluded from the results. However, this finding stresses the 312 importance of monitoring every source of potential contamination to reduce bias in MP analysis. 313

314 MP shape and polymer composition

Fibers were found in all samples, but while they comprised the minority of shapes in the pump
samples (18%), fibers were the dominant shape in the bongo samples (73%) (Fig. 2a, b). Fibers
were also the predominant shape in Manta trawls (333 µm mesh) close to Svalbard (Lusher et al.,
2015) and vertical tows with a plankton net (500 µm mesh) in East Greenland (Amélineau et al.,
2016). The most common polymer from both sample types was polyester, except for the bongo
sample at station 5 (Fig. 2c, d). This is in accordance with other Arctic studies, which found
polyester to dominate (Amélineau et al., 2016; Barrows et al., 2018; Kanhai et al., 2020, 2018;

322 Obbard et al., 2014) (Table 1). One likely source for polyester is synthetic textiles, which fits with the finding of fibers. It has been shown that textiles can release thousands of fibers during washing, 323 which subsequently end up in the wastewater (Napper and Thompson, 2016; Pirc et al., 2016). 324 Since Nuuk has no wastewater treatment, all fibers from washing effluent are released directly to 325 326 the fjord and are a highly likely source for polyester in our samples. Furthermore, polyethylene terephthalate, which is part of the polyester family, is one of the most produced and used plastics, 327 e.g., for plastic bottles and packaging material. The next most common polymers were 328 polyamide/nylon, which is used for fishing nets, polyethylene and polypropylene, which are among 329 the top 5 high-production volume plastics globally, and epoxy/phenoxy resins, which are for 330 instance applied as adhesives or coatings in boats (Andrady, 2011; Hoge and Leach, 2016; 331 332 PlasticsEurope, 2019). Overall, the polymer composition reflects important local use of plastics: fleece clothing for low temperatures, plastic packaging, fishing, and boating. 333

334 MP distribution and sources

The relative abundance of MPs between stations was similar for both methods applied (except for 335 station 4), illustrating that both methods give a representative description of the MP distribution. In 336 general, the distribution pattern followed the oceanography of the fjord, with the lowest MP 337 concentrations (68-107 MPs m⁻³) at the meltwater-impacted stations inside the fjord (stations 1, 2 338 and 3) while the MP concentration peaked after passing the major point source of the capital Nuuk 339 (Fig. 2a, b). This indicates that Nuuk is an important source for MPs in this area, as illustrated by 340 the increase in MPs along the cruise track from the inner fjord and passing Nuuk towards the sea. 341 342 Furthermore, data on wind direction and water flow within the fjord during the sampling indicated that the upper water layer was pushed out of the fjord (data not shown). This means that plastics 343 originating from Nuuk were even more likely to be found towards the outer part of the fjord. The 344 importance of local mismanagement of plastic waste in West Greenland was also confirmed by 345

346 beach litter monitoring, which reported the majority of plastic debris to originate from local sources (PAME, 2019). This underlines the importance of local anthropogenic activities for plastic pollution 347 in comparison to long-range transport with currents, which is expected to be more prominent for 348 Greenland's east coast (PAME, 2019). Airborne transport of MPs has been suggested as an 349 important pathway. A recent study found extremely high concentrations of up to 14.4.10⁶ MPs m⁻³ 350 in Arctic snow samples (Bergmann et al., 2019). This is a relevant finding with respect to the 351 present study as the Nuup Kangerlua is fed by meltwater runoff from the Greenland Ice Sheet, 352 terrestrial runoff, meltwater from sea ice, and calved glacial ice (Mortensen et al., 2013). A 353 considerable fraction of the water in the upper layers of Nuup Kangerlua is meltwater. Part of this is 354 old ice from the bottom of the glacier that is expected to be MP-free, but melted snow significantly 355 contributes to freshwater input into the fjord. With a MP contamination of snow as reported from 356 the Fram Strait, we would expect much higher concentrations in the surface waters of Nuup 357 Kangerlua, especially in the inner fjord. Our findings indicate that the MP contamination of snow in 358 West Greenland is most likely several orders of magnitude lower than reported by Bergmann et al. 359 (2019) and the main source of MPs in the fjord is of local origin rather than airborne. However, it is 360 361 important to note, once MPs enter the water column, they are also vertically exported to the seafloor/deep waters by physical and biological processes (Choy et al., 2019; Long et al., 2015; 362 Möhlenkamp et al., 2018; Peng et al., 2018; Porter et al., 2018). In fact, marine sediments in the 363 364 deep sea are a major sink for plastic pollution (Woodall et al., 2014). More research is needed to understand the distribution and fate of MPs in the Arctic ecosystem. 365

366 MP mass versus number

For the pump samples, mass concentrations of MPs were determined, which ranged from 9.3 to 51.3 μ g m⁻³, with a median of 28.8 μ g m⁻³ (Fig. 3a). The polymer composition in terms of mass differed from the particle number-based composition (Fig. 2c). Polyethylene constituted the highest 370 mass in the stations inside the fjord (stations 1 and 2), polyvinyl chloride dominated at station 3, polyester at stations 4 and 5 close to Nuuk, and polyamide was the polymer with the highest mass at 371 372 station 6, close to the open ocean (Fig. 3b). Also, the distribution of MPs between stations gave a somewhat different picture than the number-based results. While the stations from Nuuk towards 373 the open ocean (stations 4-6) still had higher concentrations than the more inward stations 2 and 3, 374 station 1, furthest inside the fjord, showed the second-highest MP abundance (Fig. 3a). This speaks 375 for the presence of a few larger (or heavier) plastic particles at that station. Plastic concentrations 376 377 are rarely reported in terms of mass for water samples, especially for small-sized MPs, even though models of plastic input and abundance in the oceans on a big scale often report mass (Cózar et al., 378 2017; Jambeck et al., 2015; van Sebille et al., 2015). Cózar et al. (2017) estimated the total load of 379 380 plastics floating on the surface of the Arctic Ocean to range between 100 and 1200 tons. The Arctic Ocean has an area of $1.4 \cdot 10^7$ km². Assuming that the surface area equals the upper 1 m of the water 381 column, the reported mass equals a mass concentration of 7-86 μ g m⁻³. Although this is a rough 382 estimate, our values fall within this range. 383

Our results illustrate that the decision of whether to report MPs in terms of mass or number will influence the picture that we get of the MP contamination. Both have their strengths and weaknesses. Mass concentrations on the one hand can be more informative about the sources and input of plastics into the environment. On the other hand, number-based concentrations are important in a biological context when considering organisms that interact with individual particles. In the best case, both should be reported. However, if only one is chosen, this choice should be based on a thorough consideration of what the results will be used for.



Figure 3. (a) Mass-based microplastic (MP) concentrations (µg m⁻³) of the pump samples and (b)
polymer composition based on estimated mass.

394

395 **Biological implications**

- 396 The dominant size fraction of MPs found in the fjord is the most available for plankton-feeding
- 397 marine invertebrates. These particles overlap in size with their natural prey, and the ingestion of
- 398 MPs between 7 and 150 µm has, for instance, been observed for different copepod species (Cole et

399 al., 2013; Sun et al., 2017; Vroom et al., 2017). The high abundance and vital role of copepods in the Arctic ecosystem illustrate their key position as an entry point for MPs into Arctic food webs. 400 401 However, compared to their phytoplankton prey, the mass of plankton-sized MPs is more than 3-4 orders of magnitude lower than that of phytoplankton (Fig. 4), which illustrates the low probability 402 403 for ingestion and impact on zooplankton. The ratio between phytoplankton and MPs depends, however, on the location. While the ratio was high in the inner fjord due to a phytoplankton bloom, 404 it was considerably lower from Nuuk towards the open ocean (stations 4-6) (Fig. 4). This means 405 that the likelihood for a plankton-feeding organism to ingest MPs differs greatly between stations, 406 depending on the phytoplankton biomass "background". Benthic invertebrates like crustaceans and 407 bivalves, as well as fish, have been found to contain MPs (Abbasi et al., 2018; Davidson and Dudas, 408 409 2016; Devriese et al., 2015; Li et al., 2018). Furthermore, studies have documented the trophic transfer of MPs (Setälä et al., 2014; Welden et al., 2018). Thus, MP pollution is of concern to 410 marine food webs. However, at the concentrations of MPs found in this study, a minor impact on 411 the Arctic pelagic food web is expected. Still, in a predicted scenario of increasing plastic pollution 412 (Lebreton et al., 2019; Lebreton and Andrady, 2019) and accelerated melting of Arctic sea ice, 413 414 increased MP pollution, together with other anthropogenic stressors, could negatively affect the 415 Arctic pelagic food web. This could be a major concern for the fishing and seafood sector in Greenland in the future, where the economy heavily depends on unpolluted marine resources 416 417 (Jacobsen et al., 2018). Therefore, it is of paramount importance to get a better understanding of the sources, distribution, and abundance of MPs in the Arctic waters, and to reduce global plastic 418 419 pollution that will end up in the Arctic.





421 Figure 4. Mass concentrations ($\mu g m^{-3}$) of phytoplankton (green), based on the fluorescence

422 measurements of the CTD, and microplastics (MPs) (grey) at the six sampling stations. The red line

423 shows the ratio of phytoplankton to MPs in terms of mass.

424 Acknowledgements

425	We thank the crew on R/V Sanna for their help during the sampling. For financial support, we
426	would like to thank the Velux foundation through the project MarinePlastic (Project no. 25084).
427	This paper was furthermore supported by the H2020 CLAIM project (Cleaning Litter by developing
428	and Applying Innovative Methods in European seas; Grant Agreement No. 774586) to TGN and a
429	research grant ("Microplastic" project no. 39610, 2019) from the Maritime DTU-Orients Fund to
430	RA.
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633 Graphical abstract

