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Greenland Sea Gyre increases microplastic pollution in the surface waters of the Nordic Seas



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HIGHLIGHTS

GRAPHICAL ABSTRACT



- · Abundance of microplastics in the East Greenland Current was 1.19 \pm 0.28 items/L
- · Abundance of microplastics in the Greenland Sea Gyre (GSG) was 2.43 \pm 0.84 items/L.
- · Microplastics in group GSG showed higher homogeneity of size, shape, and color.
- Greenland Sea Gyre increases microplastic pollution in the seawater of this sea area.

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Greenland Sea Gyre (GSG) accumulates floating microplastics in the Nordic Seas



ABSTRACT

Microplastics are ubiquitous in various ocean areas and have aroused global concern. This study investigated the abundance and characteristics of microplastic pollution in the Nordic Seas. In the sea area affected by the East Greenland Current, the abundance of microplastics was 1.19 ± 0.28 items/L, with fiber (76.1%), transparent (76.2%), and small microplastics (0.1–0.5 mm, 48.1%) being the most abundant types present. The abundance of microplastics in the cold basin affected by the Greenland Sea Gyre was 2.43 ± 0.84 items/L. Fiber accounted for 87.2% of the total microplastics, and the proportions of transparent and 0.1-0.5 mm particles were 87.6% and 63.9%, respectively. Principal component analysis (PCA) based on size spectrum data revealed that the spatial pattern of microplastics was closely related to ocean currents and the station position in the ocean current. Furthermore, scanning electron microscopy (SEM)/energy dispersive spectroscopy (EDS) analysis illustrated that the microplastics had many weathering and exfoliation sites and adsorbed heavy metals onto their surfaces. The Fourier Transform Infrared Spectrometer (FTIR) analysis showed that the microplastics in the Nordic Seas were mainly polyester and polyethylene. These results not only provide the latest data on microplastic pollution in the Nordic Seas, but also give evidence that ocean currents affect the transport of marine microplastics.

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1. Introduction

Microplastics, defined as plastics with sizes below 5 mm, are divided into two categories: primary microplastics and secondary microplastics (Arthur et al., 2009). Primary microplastics are plastic particles which serve as the feedstock in the plastic industry and as abrasives in cosmetics; while secondary microplastics are eroded from larger pieces of objects, such as car tires, textiles, and plastic containers (Napper and Thompson, 2016; Napper and Thompson, 2016). In 2015, approximately 380 million tons of plastic was produced and approximately 2-5% of that amount (7.6-19.0 million) was estimated to enter the oceans (Jambeck et al., 2015; Geyer et al., 2017). Most of these plastics flowed into the ocean through hydrodynamic systems, floated on the sea surface, and gradually degraded into microplastics due to physical, chemical and biological processes (Barnes et al., 2009). Microplastics have a high chemical stability and can remain in the environment for decades or even hundreds of years, which will lead to the accumulation of microplastics in the ocean (Cole et al., 2011).

Microplastics are widely distributed in global marine environments, including maritime spaces rarely influenced by human activities, such as Arctic deep-sea sediments, the Marina Trench, and Arctic sea ice (Bergmann et al., 2017; Peeken et al., 2018; Jamieson et al., 2019). For example, the average abundance of microplastics in near-surface waters of the polar mixed layer was 0.7 particles/m³ (La Daana et al., 2018). By contrast, microplastics with a higher abundance are usually detected in sea areas of frequent human activity. Microplastic abundance in the surface water between Bear Island and Svalbard was 2.68 \pm 2.95 items/m³ (Lusher et al., 2015); In the Yangtze Estuary of China, the microplastic abundance was up to 4137.3 \pm 2461.5 items/m³ (Zhao et al., 2014). Microplastics floating on surface waters can be easily ingested by zooplanktons (Cole et al., 2013), bivalves (Wang et al., 2019), and fishes (Ory et al., 2017). The ingestion of microplastics may cause mechanical effects (hindering mobility and clogging of the digestive tract) and physiological effects (inflammation, hepatic stress, decreased growth) (Auta et al., 2017). In addition, microplastics may enter human bodies through the food chain and cause potential implications on human health (Van Cauwenberghe and Janssen, 2014; Carbery et al., 2018). Therefore, there is an urgent need to evaluate microplastic pollution in sea areas with high capture fisheries and/or aquaculture.

As a significant site of fishery resources in the North Atlantic, the Nordic Seas occupy an important position in the global ecosystem, with a high economic and ecological value. The Nordic Seas are important feeding areas for Atlantic mackerel (Scomber scombrus), blue whiting (Micromesistius poutassou), and other pelagic fish stocks (Nøttestad et al., 2016; ICES, 2017). The Nordic Seas have three sea areas: The Greenland Sea, Iceland Sea and Norwegian Sea, and possess several ocean currents, including the Greenland Sea Gyre, East Greenland Current, and Norwegian Atlantic Current, along with numerous marine geomorphic structures (Mohns Ridge, Knipovich Ridge and Greenland Basin) (Bourke et al., 1987; Yue et al., 2019). These specific ocean currents and topographic features might accelerate the transportation and accumulation of microplastics. However, studies on microplastic pollution in the Nordic Seas are relatively rare. Therefore, the objectives of this study were to: 1) investigate the abundance, characteristics, and distribution patterns of microplastics in Nordic Seas; 2) explore the potential relationship between microplastics and ocean currents; 3) provide basic data for future microplastic research in the polar ocean.

2. Materials and methods

2.1. Sample collection

Seawater was collected from 20 stations in the Nordic Seas from the 4th to the 16th of October 2018 (Fig. 1). Nine of these stations (1-9) in a cold basin affected by the Greenland Sea Gyre and were defined as group GSG. The other 11 stations (10-20), affected by the East

Greenland Current, were defined as group EGC. In addition, three stations (1, 2, and 9) located on the periphery of the Greenland Sea Gyre were defined as group GSG-1, and the other stations (3, 4, 5, 6, 7, and 8), distributed in the interior region of the Greenland Sea Gyre, were named group GSG-2 (Fig. S1). The coordinates and measurements of temperature and salinity of each station are shown in Table S1.

Surface seawater (100 L, 10–50 cm) was drawn by a pump and directly rushed into the plankton net (Prider Instrument, Beijing) with an aperture of 0.05 mm after being filtered through a 5-mm stainless steel sieve. The filtered seawater was used to wash the plankton net three times, and the trapped particles, with a diameter of 0.05–5 mm, were collected into glass bottles and stored at 4 °C in the dark prior to analysis.

2.2. Microplastic isolation

In the laboratory, 20 mL of 30% H_2O_2 was added to the water samples and allowed to stand for 24 h at room temperature to dissolve the organic matter in accordance with the method presented by Estahbanati and Fahrenfeld (2016) and Wang et al. (2017). The plastic particles were then suspended using saturated ZnCl₂ solution (1.6 g /cm³) overnight (Wolff et al., 2019). The suspended seawater was filtered successively through stainless-steel sieves with apertures of 2 mm, 1 mm, 0.5 mm, and 0.1 mm, respectively. The trapped particles were washed into petri dishes with 0.45-µm-filtered double-distilled water and placed in a 60 °C incubator for 24 h.

To minimize microplastic loss, the suspension, filtration and washing processes were repeated three times. Moreover, the methods of La Daana et al. (2017) were adopted to avoid microplastic contamination, including the use of 0.45-µm filtered liquids, the covering of all containers, and the wearing of exclusive cotton laboratory coats and nitrile gloves. The observations were carried out in a closed room, which was cleaned before every use. Three blank controls (0.45-µm-filtered double-distilled water) were run through the entire process to test whether the count was polluted by aerial microplastics. No microplastics larger than 0.1 mm were detected in the blank controls.

2.3. Identification of microplastics

The petri dishes were placed under a stereoscopic microscope (JSZ6, Nanjing Jiangnan Novel Optics Co., Ltd., China) for preliminary observation using the identification criteria of Nor and Obbard (2014). According to the mesh sizes of the stainless-steel sieves, microplastics were divided into four size ranges: 2-5 mm, 1-2 mm, 0.5-1 mm and 0.1–0.5 mm. In addition, microplastics were categorized into five shapes (lines, fibers, granules, films and fragments) and six colors (transparent, black, blue, yellow, green, and red). Fibers were slender and greatly elongated; while lines were thick, short, and straight. Fragments had smooth surfaces and particularly jagged edges, and granules comprised regular pellets and irregular solid particles. Randomly selected microplastics (n = 200, size ≥ 1 mm) were grinded with potassium bromide in an agate mortar and placed in an infrared spectrometer (NICO-LET iS10) for FTIR analysis (Sun et al., 2017). The analyses were performed in reflection mode in the range of $400-4000 \text{ cm}^{-1}$, with 32 scans at a resolution of 4 cm^{-1} . After automatic baseline calibration, the obtained spectra were compared with the infrared spectra of different plastic materials in the references to identify the microplastic components. The microplastics were then classified into the following types: polyethylene, polypropylene, polystyrene, polyamide, polyvinyl acetate, and polymethyl methacrylate.

2.4. Surface morphology and elemental analysis

Randomly selected microplastics (n = 24) were fixed with evaporated gold, and their surface morphology was observed using a scanning electron microscope (SEM, model s-3400n, Japan). The magnification of the SEM was 200–4000×, and the acceleration voltage was 20.0kv. The



Fig. 1. Map of the studied 20 stations in the surface water of Nordic Seas. The area enclosed in the red rectangle represents the sampling region. Stations 1–9 were distributed in the cold basin that affected by the Greenland Sea Gyre, and stations 10–20 were affected by the East Greenland Current.

qualitative elemental composition of microplastics was analyzed by energy dispersive X-ray spectroscopy (EDS; EMAX, Japan) (Wang et al., 2017).

2.5. Statistical analysis

Multivariate analyses were conducted using the PRIMER v 6.1 package (Clarke and Gorley, 2006) and PERMANOVA+ for PRIMER (Anderson et al., 2008). The spatial pattern of microplastics in the sampling area was summarized via principal component analysis (PCA) based on logarithmic transformation/normalized size spectrum data (Jiang et al., 2014). The differences among the groups discriminated by PCA were tested by PERMANOVA (Anderson et al., 2008; Jiang et al., 2011). Oneway ANOVA was applied to assess the differences of microplastic abundances among the groups, followed by multiple pair-wise comparisons between each pair of groups using the statistical software IBM SPSS v22.0.

3. Results

3.1. Microplastic abundance

The average abundance of microplastics in group GSG was 2.43 \pm 0.84 items/L, with the largest number of microplastics in stations 6

and 4 (3.74 ad 3.72 items/L, Fig. 2, S2). While microplastics in group EGC had an average abundance of 1.19 ± 0.28 items/L, and stations 13 and 14 had the lowest microplastic abundance (0.88 and 0.8 items/L). Six stations in group GSG had microplastic abundances of >2 items/L, while the microplastic abundances in almost all the stations in group EGC (except for station 17) were below 1.5 items/L (Fig. S2).

The microplastic abundance in group GSG-2 was significantly higher than in GSG-1 and EGC (P < 0.05, Fig. 3). Furthermore, GSG-2 had the highest percentages of 0.1–1.0 mm size range, fiber, and transparent microplastics (Fig. 3, Table S2).

3.2. Microplastic characteristics

3.2.1. Size distribution

In group GSG, 63.9% of the total microplastics were 0.1–0.5 mm, and 1–2 mm microplastics were present in the lowest proportion (8.4%) (Fig. 4a). By contrast, microplastics in the size range of 0.1–0.5 mm accounted for 48.1% of the total particles in group EGC, followed by 0.5–1 mm (22.7%), 2–5 mm (16.6%), and 1–2 mm (2.59%) (Fig. 4d). High percentages of 0.1–0.5 mm microplastics were observed in station 1 (88%), station 2 (75%), and station 9 (75%), while station 18 had the highest percentage of 2–5 mm microplastics (30%, Fig. S3).



Fig. 2. Abundance of microplastics in the seawater collected from 20 stations (area enclosed in the red rectangle) in the Nordic Seas. Hydrographic maps in the Nordic Seas: EGC, East Greenland Current; GSG: Greenland Sea Gyre; NwAC: Norwegian Atlantic Current.

3.2.2. Shape distribution

Fibers were the dominant microplastic shapes, accounting for 87.16% in group GSG and 76.13% in group EGC (Fig. 4b, e). In group EGC, the percentages of microplastics in the shapes of lines (8.39%), granules (1.83%), films (2.44%) and fragments (11.21%) were higher than those in group GSG. In addition, the percentages of fiber microplastics in stations 4, 5, 6 and 8 were higher than 90%, while

>20% of the microplastics were fragments in stations 14, 15 and 16 (Fig. S4).

3.2.3. Color distribution

Most of the microplastics in the two groups were transparent, with 87.6% in group GSG and 76.2% in group EGC (Fig. 4c, f). The percentage of blue particles, the second most prominent microplastics, in group



Fig. 3. Differences in abundance, and size, shape, and color compositions of microplastics in the three sea areas, including the peripheral area of the Greenland Sea Gyre (GSG-1), the interior region of the Greenland Sea Gyre (GSG-2), and East Greenland Current (EGC). **P* < 0.05.

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Fig. 4. Composition of different sizes, shapes, and colors of microplastics in the sweater collected from group GSG (a-c) and group EGC (d-f) in the Nordic Seas.



Fig. 5. Microplastic images under a scanning electron microscope: a, film; b, fiber; c, line; d, fragment. Scale bar (bottom right corner) = 100 µm.

GSG and group EGC were 6.6% and 16.17%, respectively. In group EGC, nine stations had >15% of blue microplastics, but no station in group GSG recorded such a high percentage of blue microplastics (Fig. S5).

3.3. Surface morphology and chemical composition

Different forms of microplastics, such as transparent film, red fiber, blue fragment, blue line, yellow granule, and yellow film, were observed under the stereoscopic microscope (Fig. S6). SEM analysis showed that the surface of thin film microplastics contained a discernable stripping phenomenon (Fig. 5a). Fiber microplastics encompassed large twists and curves (Fig. 5b), while linear microplastics exhibited larger widths and lower bends (Fig. 5c). For fragment microplastics, numerous cracks were irregularly distributed on their smooth surfaces (Fig. 5d).

High amounts of carbon and oxygen were detected on fiber, line, and film microplastics by EDS analysis (Figs. S7–S9). Other elements, such as chlorine, silicon, calcium, and zinc, were also found on the surfaces of the microplastics.

3.4. Microplastic composition

FTIR analysis identified at least seven plastic components, including polyester (35%), polyethylene (PE, 26%), polypropylene (PP, 8%), polystyrene (PS, 2%), polyvinyl acetate (PVAc, 6%), polyamide (PA, 5%), and three other types (Fig. Error! Reference source not found. and Fig. S10). However, many fibers in the samples were identified as cellulose (9%) by the following FTIR verification (Fig. 6).

3.5. The spatial pattern in size spectrum of microplastics

Principal component analysis (PCA) based on the microplastic abundance data from 20 sampling stations (Fig. 7). The first PCA axis in the plot showed that the total variability was 59.2%, which separated the sampling stations in groups GSG-1 (station 1, 2, 9) and EGC (station 10–20) (to the left of the plot) from the sampling stations in group GSG-2 (to the right of the plot). The second axis demonstrated 27.8% of the environmental variability that discriminated the EGC group from the GSG-1 and GSG-2groups. A permutational multivariate analysis of variance (PERMANOVA) test revealed significant differences among sample clouds from the three groups (pseudo-F = 9.010, P = 0.001, Table S3).

4. Discussion

This study investigated microplastic pollution in the Nordic Seas and found that ocean currents, especially the Greenland Sea Gyre, substantially influence the distribution and characteristics of microplastics in this sea area. The average abundance of microplastics in surface seawaters of the Nordic Seas was 1.76 items /L, which was higher than the abundances recorded in the sub-surface waters (6 m below surface) of Northeast Greenland (2.4 items/m³) in August 2015 (Morgana et al., 2018) and the surface waters of the Svalbard archipelago in June 2014 (0.34 items/m³, Lusher et al., 2015). Due to the lack of standardized methods, this research, including two prior studies, have adopted different sampling methods (pump and trawl sampling) and microplastic isolation procedures. Therefore, variations in the results obtained from this study in comparison to previous studies are possibly attributed to these



Fig. 6. FTIR spectra for representative microplastics collected in the Nordic Seas: PA, polyamide; PE, polyethylene; PET, polyethylene terephthalate; PMMA, polymethyl methacrylate; PP, polypropylene; PS, polystyrene.



Fig. 7. Principal component analysis (PCAs) of spatial pattern in size spectra of microplastics based on sampling stations; GSG, Greenland Sea Gyre; EGC, East Greenland Current.

different methods (Table 1). In addition, Arctic sea ice is considered as an important temporal sink for microplastics and the concentration of microplastics in the ice core was up to 1.2×10^7 items/m³ (Peeken et al., 2018). Obbard et al. (2014) warned that global warming would release microplastic legacy frozen in Arctic Sea ice. Thus, the increase of microplastics in our study might be partly related to the flow of melting sea-ice during these years.

The Nordic Seas have two main ocean currents, the East Greenland Current (EGC) and the Greenland Sea Gyre (GSG). The EGC is formed by arctic water flowing southward along the Fram Strait and cold melting water from Greenland; the GSG is the counterclockwise circulation in the Greenland Basin formed by a branch of the Norwegian Atlantic Current (NwAC), which flows northeasterly along the Atlantic coast of Norway and is considerably warmer and saltier than the Arctic Ocean (Orvik and Niiler, 2002; Rossby et al., 2009; Raj et al., 2015). This study compared the abundance and characteristics of microplastics in these two areas and found that the microplastic abundance in the GSG group was approximately two-fold of that in the EGC group. Moreover, the stations with the highest microplastic abundances were distributed in the center of the GSG. Once plastic or microplastics enter the ocean, they can be extensively distributed by wind and water currents (Eriksen et al., 2013; Do Sul and Costa, 2014). For example, Isobe et al. (2017) reported that the Antarctic Circumpolar Current spread significant concentrations of microplastics in the Southern Ocean, and oceanic circulation models suggest that five subtropical ocean gyres might be the possible accumulation regions of microplastics (Maximenko et al., 2012; Lebreton et al., 2012). In this study, high levels of microplastics were detected in the Greenland Sea Gyre, which supports these model results. Most microplastics in the Nordic Seas were below 0.5 mm, fiber, and transparent, which are similar to the characteristics of microplastic pollution in other sea areas (Hidalgo-Ruz et al., 2012; Zhang et al., 2017). Moreover, the microplastics in the Greenland Sea Gyre showed higher percentages of small size range (0.1–0.5 mm, 63.9%), fiber shape (87.2%) and transparent color (87.6%) microplastics compared to those in the East Greenland Current. Moreover, PCA result discriminated the sample clouds from GSG and EGC based on the abundance and characteristics of microplastics, and a significant difference was found between the two regions (P < 0.05). Therefore, these results revealed that ocean circulation not only increased microplastic abundance, but also homogenized the composition of microplastics in their size range, shape, and color category. The low levels of microplastics in the EGC might be related to the effects of arctic water from the Fram Strait and the fresh water from the inaccessible East Greenland, which is minimally affected by anthropogenic activities (Rigét et al., 2019). Conversely, microplastics in the GSG are predominately derived from the NwAC, which originates from the low-latitude North Atlantic Current, and the high temperatures may accelerate the degradation rates of larger microplastics, breaking them down into small-size particles. Meanwhile, areas in lower latitudes are exposed to higher intensities of ultraviolet radiation, which may serve as an important factor in speeding up the decomposition rate (Andrady, 2011; Bonhomme et al., 2003). Therefore, many environmental and human factors should be considered to accurately evaluate microplastic pollution in a specific sea area.

Table 1

Comparison of microplastic abundance (items/m³) in the seawaters in the sea areas around the Nordic Seas.

Study area	Sampling time	Depth (m)	Sampling method	Mesh size (µm)	Abundance (items/m ³)	References
Arctic Central Basin	September. 2016	8.5 m	Universal II Series Pump	250 µm	0.7 (median)	La Daana et al., 2018
Arctic waters south and southwest of Svalbard, Norway	June. 2014	16 cm (surface); 6 m (sub-surface)	Manta net	333 μm	$\begin{array}{l} 0.34 \pm 0.31 \text{ (surface);} \\ 2.68 \pm 2.95 \text{ (sub-surface)} \end{array}$	Lusher et al., 2015
North Pacific Subtropical Gyre (NPSG)	2009-2010	Surface water	Standard manta net	333 µm	0.12	Goldstein, 2012
Northeast Greenland	August. 2015	6 m	Water intake with rubber sealings	80 µm	2.4 ± 0.8	Morgana et al., 2018
Fram Strait and Central Arctic	Spring 2014-Summer 2015	Sea ice cores	-	-	$(1.1 \pm 0.8) imes 10^6$ to $(1.2 \pm 1.4) imes 10^7$	Peeken et al., 2018
Arctic Sea	-	Sea ice cores	Band saw	0.22 µm	38-234	Obbard et al., 2014
Greenland Sea	August. 2014	Surface water	Towing net	100 µm	2.38 ± 1.11	Amélineau et al., 2016
South Pacific subtropical gyre	March. 2011	Surface water	Manta trawl	333 µm	26,898 items $\rm km^{-2}$	Eriksen et al., 2013:
Greenland and Barents seas	_	Sub-surface water	-	-	6.3×10^3 items \cdot km ⁻² (median)	Cózar et al., 2017
Nordic Seas	October. 2018	Surface water	Pump	100 µm	800-3740	This study

PCA results demonstrated that the GSG samples could be further divided into two groups, which were situated at the center and at the edge of the Greenland Basin, respectively. Kostigen and Magazine (2008) reported that the accumulation of plastic marine debris in the center of the North Pacific Gyre might be influenced by the "circular effect", but the objectives of his study are limited to large-sized plastics and other kinds of particles. This study found that microplastic abundance in the center of the GSG is significantly higher than that in the edge, confirming that microplastic pollution in Nordic Seas follows a similar spatial pattern. Furthermore, surface morphology, chemical composition and plastic types were analyzed in this study. The results of SEM analysis showed that the surface of film microplastics were like onion skins, and the fragment microplastics had many irregularly cracks, an obvious stripping and weathering appearance, which may be attributed to the ageing of plastics (Hüffer et al., 2018; Kedzierski et al., 2018). The linear and fiber microplastics contained high amounts of carbon, oxygen and chlorine, which proved that they were non-biological organics subsequent to treatment with hydrogen peroxide (Ding et al., 2019). Low amounts of Zn were also detected in the microplastics, particularly in comparison to heavy metals detected in the microplastics from the costal sea area (Rochman et al., 2014; Wang et al., 2019). Thus, the absorbed chemicals on microplastics might be a useful indicator of pollution in marine environments. Polyester and PE were found to be the most common plastic types in the Nordic Seas, and PP, PA, PS were also found. The total proportion of Polyester and PE was >50%, which was consistent with the results of Norwegian coast (Bråte et al., 2016), and the high polyester percentage result was similar to studies conducted on Arctic Sea ice (Obbard et al., 2014) and in the Arctic Central Basin (La Daana et al., 2018). Polyester is widely used in clothing, and the washing of clothing has been suggested as an important source of microplastics (Hernandez et al., 2017). PE is the most widely used type of plastic throughout the world and one of the mass-manufactured polymers found in terrestrial environments (Kalogerakis et al., 2017). Thus, finding substitutes for plastics and decreasing plastic debris entering the ocean might reduce the rate of microplastics accumulation. Previous studies reported that cellulose fibers could not be easily excluded during microplastic isolation procedures (Song et al., 2015; Remy et al., 2015). In our study, approximately 9% of the microplastics were finally found to be cellulose. Lusher et al. (2015) also reported that 30% of fibers were identified as cellulose via FTIR analvsis. However, it is not suitable to arbitrarily remove these particles because cellulose has an almost identical FTIR spectra to rayon, a semisynthetic polymer (Lusher et al., 2014). To resolve this issue, more accurate separation and identification techniques should be considered.

5. Conclusion

This study not only provided basic data on the abundance, size, shape, color, and distribution of microplastics in Nordic Seas, but also found that the spatial pattern of the microplastic size spectra in surface seawaters is closely related to ocean currents. The Greenland Sea gyre could increase microplastic pollution in this sea area, especially towards the center of the Ocean Circulation. Therefore, the impact of the ocean currents on the distribution of microplastics should not be ignored in future studies, and the use of size spectrum patterns, which are might be a useful tool to assess microplastic pollution.

Declaration of competing interest

We declare that we have no financial and personal relationships with other people or organizations that can inappropriately influence our work, there is no professional or other personal interest of any nature or kind in any product, service and/or company that could be construed as influencing the position presented in, or the review of, the manuscript entitled "Greenland Sea Gyre increases microplastic pollution in the surface waters of the Nordic Seas".

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi. org/10.1016/j.scitotenv.2019.136484.

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