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Anthropogenic particles (including microfibers and microplastics) in marine sediments of the Canadian Arctic

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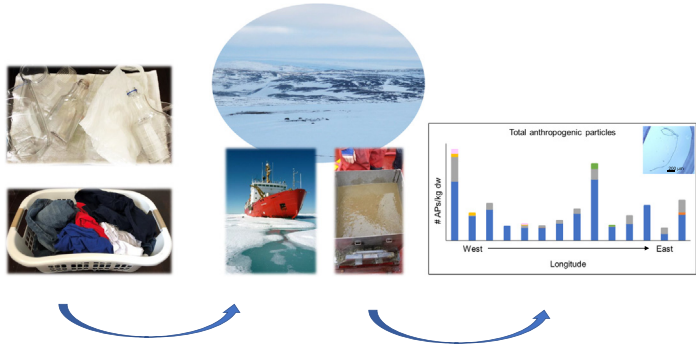
HIGHLIGHTS

- Concentrations and composition of APs were identified from Canadian Arctic marine sediments.
- Sediments contained high concentrations of APs, dominated by microfibers.
- Anthropogenic cellulose comprised the greatest proportion of microfibers.
- High AP concentrations suggest potential for both long-distance and local sources.

GRAPHICAL ABSTRACT



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ABSTRACT

We report the first Canadian Arctic-wide study of anthropogenic particles (APs, >125 μm), including microfibers (synthetic, semi-synthetic and anthropogenically modified cellulose) and microplastics, in marine sediments from 14 sites. Samples from across the Canadian Arctic were collected between 2014 and 2017 from onboard the CCGS *Amundsen*. Samples were processed using density separation with calcium chloride ($CaCl_2$). APs >125 μm were identified and a subset (22%) were characterized using Raman spectroscopy. Following blank-correction, microfiber numbers were corrected using Raman data in a novel approach to subtract possible "natural" cellulose microfibers with no anthropogenic signal via Raman spectroscopy, to estimate the proportion of cellulose microfibers that are of confirmed anthropogenic origin. Of all microfibers examined by Raman spectroscopy, 51% were anthropogenic cellulose, 11% were synthetic polymers, and 7% were extruded fibers emitting a dye signal. The remaining 31% of microfibers were identified as cellulosic but could not be confirmed as anthropogenic and thus were excluded from the final concentrations. Concentrations of confirmed APs in sediments ranged from 0.6 to 4.7 particles g^{-1} dry weight (dw). Microfibers comprised 82% of all APs, followed by fragments at 15%. Total microfiber concentrations ranged from 0.4 to 3.2 microfibers g^{-1} dw, while microplastic (fragments, foams, films and spheres) concentrations ranged from 0 to 1.6 microplastics g^{-1} dw. These concentrations may exceed those recorded in urban areas near point sources of plastic pollution, and indicate that the Canadian Arctic is a sink for APs, including

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anthropogenic cellulose fibers. Overall, we provide an important benchmark of AP contamination in Canadian Arctic marine sediments against which to measure temporal trends, including the effects of source reduction strategies and climate change, both of which will likely alter patterns of accumulation of anthropogenic particles.

1. Introduction

Remote Arctic regions in Canada are comprised of unique ecosystems that support the wellbeing of Indigenous peoples. These regions are sentinels for global environmental change, and have been experiencing the effects of climate change at twice the rate than the global average (Mueller et al., 2009; Woodward et al., 2010; Box et al., 2019; Bush and Lemmen, 2019). Additionally, northern environments for decades have been accumulating the burden of global contaminants produced in more southerly locations (Macdonald et al., 2000; Lohmann et al., 2007; Carrizo and Gustafsson, 2011). Recently, anthropogenic particles (APs) have also been observed across the Arctic (e.g., Bergmann et al., 2017; Kanhai et al., 2018a; Huntington et al., 2020; Table S1). Herein, we define APs as particles found in the environment of manufactured origin, as the sum of microplastic (plastic particles <5 mm), and microfiber particles, including synthetic, semi-synthetic, and non-synthetic particles or fibers of anthropogenic origin (Collard et al., 2018; Athey et al., 2020). APs are an emerging contaminant class, increasing the threat to Arctic wildlife and human populations through consumption, and may adversely impact aquatic biota and food webs through ingestion, entanglement, and smothering (Rochman et al., 2016; Provencher et al., 2019). For this reason, the Arctic Monitoring and Assessment Programme (AMAP) has added marine plastics and microplastics to the list of Chemicals of Emerging Arctic Concern (AMAP, 2017), and the G7 has adopted the Canada-led initiative "Ocean Plastics Charter", with the goal of moving towards more resource-efficient and manageable production, usage, and disposal of plastics globally. Baseline data are needed to assess the future effectiveness of these policy directives.

Microplastics can be classified as either primary or secondary origin (Duis and Coors, 2016). Primary origin microplastics are produced either as microspheres or pellets for the purposes of industrial applications, plastic production, or use in personal care products, although many countries have now banned this latter use. Secondary origin microplastics form from the degradation and/or fragmentation of larger plastic objects (Wallace and Arthur, 2014). Tire wear particles, which are small black rubber particles found in the environment (Sommer et al., 2018), and cellulosic foams and fragments of anthropogenic origin are included here under the term "microplastics". Here, we consider microfibers that include anthropogenically modified natural (e.g. cotton, hemp), semi-synthetic (e.g., rayon) and synthetic materials (e.g. polyester, nylon, acrylic) (Fontana et al., 2020; Athey et al., 2020). Microfibers are primarily released from the shedding of textiles. APs contain intentionally added chemicals. For example, plastics are produced with plasticizers, colorants, stabilizers, and flame retardants which can be released into the surrounding environment. APs can also sorb pollutants from the environment, including persistent organic pollutants (POPs), notably dichlorodiphenyltrichloroethanes (DDTs), polychlorinated biphenyls (PCBs), and hexachlorocyclohexanes (HCHs) (Colabuono et al., 2010; Van et al., 2012; Koelmans et al., 2014), and metals (e.g. lead, cadmium, and nickel) (Rochman et al., 2014). Textile additives including dyes, fillers and chemical treatments can constitute an important portion of microfiber dry masses (Xue et al., 2017). Therefore, in addition to entanglement, smothering, and nutritional depletion from ingestion of APs, toxicity of ingested APs is a cause for concern. Moreover, APs can act as transport media for invasive species to the Arctic (Gregory, 2009; Tutman et al., 2017).

APs have been identified in most environmental compartments of the circumpolar Arctic (Table S1), with studies recording both microplastics and microfibers in Arctic snow (Bergmann et al., 2019), sea ice (Obbard et al., 2014; Peeken et al., 2018; Geilfus et al., 2019; Kanhai et al., 2020), Arctic surface waters (Lusher et al., 2015; Cózar et al., 2017; Kanhai et al., 2018b; Huntington et al., 2020), sub-surface waters (Kanhai et al., 2018b; Peeken et al., 2018; Morgana et al., 2018), and deep-sea sediments (Woodall et al., 2014; Bergmann et al., 2017; Kanhai et al., 2018a; Mu et al., 2019; Huntington et al., 2020; Athey et al., 2020). APs in Arctic regions are often found at elevated concentrations (Barrows et al., 2018), despite these locations being at great distance from confirmed major sources including industry, urban centres, and agriculture (e.g. Baldwin et al., 2016; Dris et al., 2016; Piehl et al., 2018; Liu et al., 2019), suggesting the importance of long-distance transport. Important local sources, however, have also been documented in analyses of APs in wastewater, sewage effluent and near-shore marine environments in Svalbard (Granberg et al., 2019). Shipping routes are concentrated at the margins of the Canadian Arctic Archipelago; this may also be an important local source of APs, particularly as the frequencies of ship visits are increasing as sea ice declines (Fiske, 2018).

Thus, sources and transport mechanisms of APs to Arctic regions are not yet fully understood although multiple lines of evidence point to the importance of long-distance transport. Analyses of glaciers (Ambrosini et al., 2019) as well as wet and dry deposition in alpine regions (Allen et al., 2019) confirm that microplastic particles up to 300 μm in diameter and microfibers up to 750 μm in length are readily entrained in global air masses and deposited at distance from sources. Global simulations of atmospheric transport of microplastic particles indicate high transport efficiencies through the atmosphere to remote regions, with the Arctic highlighted as particularly susceptible to receipt of microplastic pollution by this mechanism (Evangeliou et al., 2020).

APs may also undergo long-range transport through ocean currents (Lusher et al., 2015), as these flows transport water and suspended sediment-bound APs northward, providing a conveyer of anthropogenic debris from tropical and temperate oceans to northern marine systems (Cózar et al., 2017). During transit, high-density APs will eventually sink through the water column and accumulate in marine sediments, while other polymers (e.g. polypropylene (PP) and polyethylene (PE)) which are less dense than sea water, may accumulate a biofilm and/or colonizing algae and invertebrates, increasing their density and causing otherwise light particles to also accumulate in the sediment sink (Woodall, 2015; Van Melkebeke et al., 2020; Wu et al., 2020). Ross et al. (2021) acquired 71 near-surface seawater samples across the European and North American Arctic and found that there was an east-to-west shift in infra-red signatures of polyester microfibers, suggesting potential weathering of fibers away from the source. This study also documents widespread distribution of microfibers in the remote Arctic, and the authors proposed that microfibers are transported to the eastern Arctic Ocean via Atlantic Ocean inputs and/or atmospheric transport from the South (Ross et al., 2021). Yakushev et al. (2021) supported the conclusions of Ross et al. (2021) for long-distance transport of APs from Atlantic Ocean currents, as they found surface waters of Atlantic origin and Great Siberian Rivers discharge were the greatest sources of microplastics in the Eurasian Arctic.

Riverine discharge to the Arctic is high and is a source of other chemical contaminants to the Arctic (Panagopoulos-Abrahamsson et al., 2020; Sühring et al., 2021). This is a third possible mode of long-distance transport of APs to Arctic marine sediments. Koutnik et al. (2021) report higher microfiber concentrations in coastal sediments than in sediments of inland water bodies, suggesting long-distance transport of microplastics from inland locations to terrestrial boundaries including estuaries and near-shore environments. In addition to

atmospheric and aquatic transport, microplastics can be transported to the Arctic by other vectors, such as migratory seabirds and other marine biota (Amelineau et al., 2016; Provencher et al., 2018). This diversity of potential sources highlights the urgent need for monitoring programs and policies to be implemented in Arctic countries to address the spread and sources of marine plastic litter (Linnebjerg et al., 2021).

Recent research indicates that anthropogenic microfibers are the dominant AP in most Arctic environmental matrices (Lusher et al., 2015; Barrows et al., 2018; Reed et al., 2018; Huntington et al., 2020; Ross et al., 2021; Table S1). Clothes laundering is likely a major source (Browne et al., 2011; Fontana et al., 2020; Athey et al., 2020). Most studies have focused on synthetic fibers (e.g. Ballent et al., 2016; Kanhai et al., 2018b). However, anthropogenically modified cellulose (AC) microfibers (e.g. from cotton-based textiles) are relatively persistent and also accumulate in the environment (e.g., Sanchez-Vidal et al., 2018; Athey et al., 2020). When included, AC microfibers have been shown to contribute significantly to microfiber abundance (e.g. Lusher et al., 2015; Table S1). To date, few Arctic studies have documented natural microfibers such as AC fibers (e.g. Athey et al., 2020; Huntington et al., 2020; Kanhai et al., 2020; see Table S1 for details). We argue that although these types of microfibers are derived from natural materials (e.g., cotton, hemp), they should not be over-looked. Textiles comprised of 100% natural fibers (e.g., 100% cotton) may contain up to 27% added chemicals by weight (Lacasse and Baumann, 2004; Xue et al., 2017). Therefore, these 'natural' microfibers need to be considered in risk assessments along with synthetic microfibers.

Research is now emerging showing the accumulation of APs in Canadian Arctic sediments; Huntington et al. (2020) measured concentrations of microplastics and microfibers in Hudson Bay and the Eastern Canadian Arctic and found that 90% of sediment samples contained APs. The study found 57% of APs were microfibers, most of which (52%) were cellulosic. Athey et al. (2020) reported that 86% of APs in sediments from the Canadian Arctic Archipelago were microfibers, of which 51% were AC microfibers. Further, they reported that 20% of all microfibers were identified as indigo denim.

This study extends the work of Athey et al. (2020) who reported aggregated (average) values of APs, AC microfibers and indigo denim microfibers from the 14 sites examined here in detail. Understanding the presence, accumulation, and behaviour of AP contamination in the Canadian Arctic is crucial, considering the implications for the health and wellbeing of Indigenous peoples and Arctic food webs (AMAP, 2017). Increasing our understanding is also particularly relevant given the recent and projected changes in the region due to climate change, including changes to ocean currents and sea ice, and related increases in accumulation of APs (Obbard et al., 2014; Welden and Lusher, 2017). Herein, we assessed the spatial extent, concentrations, and composition of APs (>125 μm) in marine sediments across the Canadian Arctic Archipelago as indicators of decadal accumulation, and evaluated spatial trends in AP distribution. We hypothesize that APs, including microplastics and microfibers, accumulate to high concentrations in the Canadian Arctic, as is observed with chemical contaminants, and that transport mechanisms common to chemical contaminants may also explain AP accumulation patterns in the Canadian Arctic.

2. Materials and methods

2.1. Study site and field collection

Surface marine sediments were sampled opportunistically from 14 sites across the Canadian Arctic Archipelago (Fig. 1). These sites and sampling procedures are presented by Athey et al. (2020). Briefly, sampling occurred during summer (May to September) as part of yearly ArcticNet expeditions in collaboration with the Northern Contaminants Program on board the CCGS *Amundsen* in 2014, 2015, and 2017. Ocean bottom depths ranged from 40 to 1502 m (Table 1). Samples were collected during periods of open-water with limited ice coverage, using a Reineck-type spade-foot corer with 0.25-m^2 surface area that penetrates ~40 cm and allows for the collection of undisturbed surface sediments. The top 1–5 cm were collected for analysis by scraping the surface layer or from push core subsamplers (Table 1). Samples were stored in glass jars (pre-cleaned with soap and water and baked at 400 °C overnight) with Teflon-lined lids at −20 °C until analysis.

2.2. Sample processing

Wet sediments were dried at 55 °C to constant mass to obtain total dry mass. All of the available sediment from each site was used in the analysis; sample masses ranged from 9 to 62 g dry weight (dw) per sample (Table 1). Following drying, samples were wet sieved using 500-μm and 20-μm stainless steel sieves by gently washing with reverse osmosis (RO) water (See Fig. S1 for methods flow chart). The ≥500 μm and 20–500 μm fractions were retained and underwent density separation in a $CaCl_2$ solution (density of 1.4 g cm^{-3}), which was filtered prior to use through a 47-mm nucleopore polycarbonate filter with a 10-μm pore size to remove possible impurities.

Separation was achieved in two stages: first for 2 h and second for 24 h. First, sediments were transferred from sieves into glass beakers using the $CaCl_2$ solution, stirred vigorously for 2 min, covered and allowed to settle for 2 h. The less dense portion of the sample was then poured from the beaker into a separatory funnel and shaken vigorously for 2 min. The sides of the separatory funnel were then rinsed with $CaCl_2$ solution, and the sample allowed to settle for 24 h (density separation stage 2). The sample was then drained from the separatory funnel in two phases, first the dense and then the less dense. The less dense portion from the separatory funnel was rinsed with RO water and size fractioned using 500-μm, 355-μm, 125-μm, and 20-μm sieves to remove the $CaCl_2$, and for ease and accuracy of picking and identification. The sample was then washed using RO water into glass petri dishes for each size fraction for further analysis.

2.3. Physical and chemical identification of anthropogenic particles

Only the >125 μm fractions were included in these analyses due to difficulties with manual handling and spectroscopic analysis of particles <100 μm. Samples were examined in a water-filled glass petri dish using a Zeiss Stereo Microscope at 25× to 80× magnification. All identified APs were removed from the dish, placed on double-sided tape in another glass petri dish, and given a particle ID number. All particle picking was done by a single analyst. APs were placed into one of six categories based on morphologies (Rochman et al., 2019; Fig. S2): fibers, fiber bundles, films, foams, fragments (including tire wear particles), or spheres, and were also classified based on colour. Finally, all picked particles were photographed and measured using Zeiss Zen software for digital microscopy.

A subset of APs was chemically characterized using a HORIBA XPlora Plus micro-Raman spectrometer (New Jersey, USA) with LabSpec 6 spectroscopy suite software and 100× objective. The micro-Raman was equipped with a 785-nm (range 50–2000 cm^{-1}) and 532-nm (range 50–4000 cm^{-1}) laser (Table S3). The subsampling strategy was to analyze a random subset totalling 10% (rounded up to the nearest whole number) of APs in each colour category (e.g. blue fiber, red fiber, clear fragment, etc.) per sample (Grbić et al., 2020). If Raman spectra indicated that a particle was natural, it was not counted towards the 10% target for Raman characterization in each colour category. This resulted in successful Raman characterization for 22% of all APs for the sample set (183 of 810 particles). Spectra were matched to standard materials catalogued in the spectral databases of both the Bio-Rad KnowItAll Spectral Library (e.g., Bio-Rad Sadtler, HORIBA Scientific and Wiley databases) and the Spectral Library of Plastic Particles (SLoPP and SLoPP-E) (Munno et al., 2020). We did not set a specific criterion for the hit quality index (HQI), although this is common in microplastic

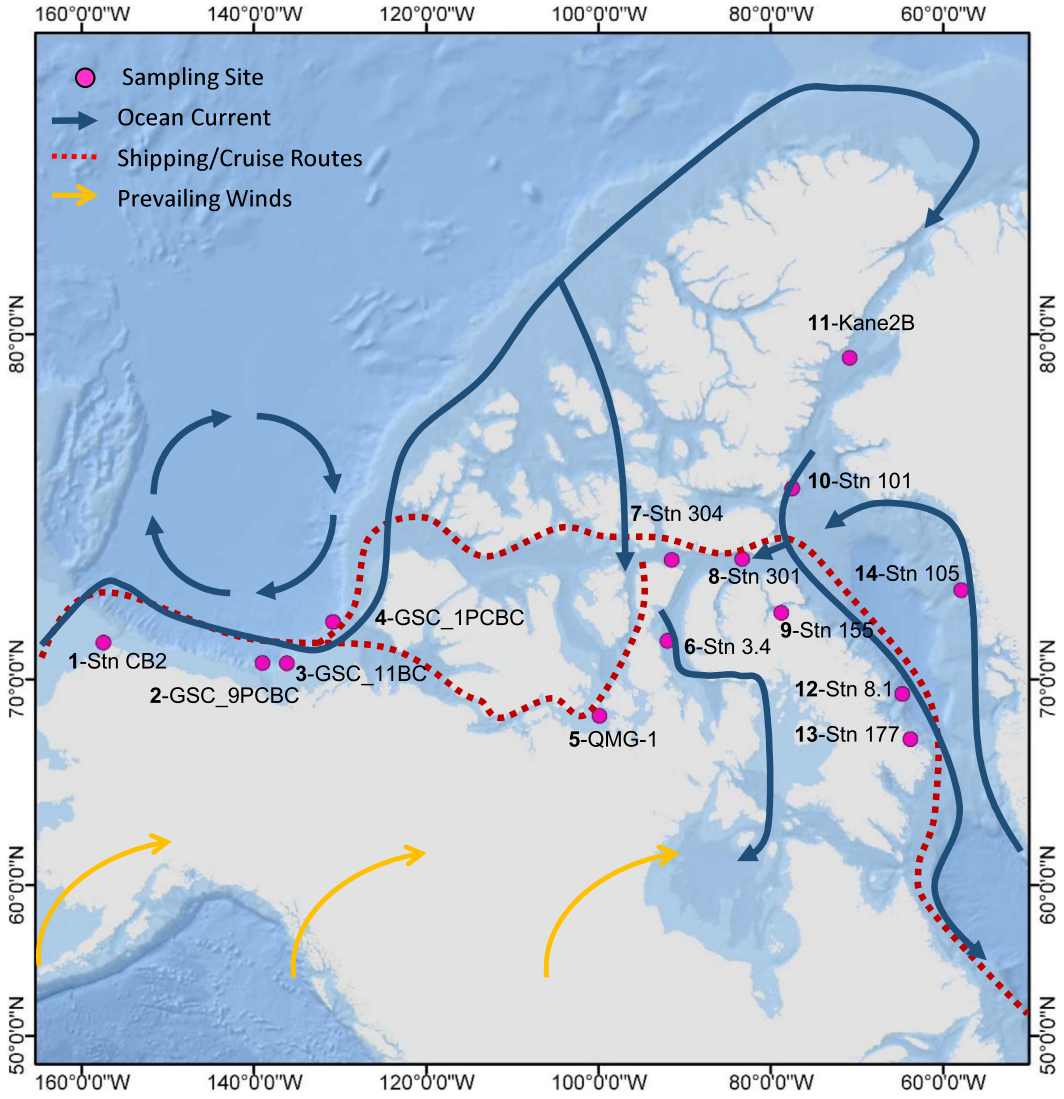

Fig. 1. Map of marine sediment sampling sites across the Beaufort Sea and the Canadian Arctic Archipelago. Site numbers and station codes are labelled for each site.

analysis. Peak intensities can differ with different instrument conditions (e.g., laser excitation) used to analyze the samples and those used to analyze reference materials, ultimately affecting HQI (Park et al., 2017). Therefore, while HQI was taken into consideration, the best quality match was visually determined based on intensity and alignment of peaks compared to standard spectra. All suspected natural particles, identified either visually or by Raman spectroscopy as non-anthropogenic, were removed from the final data set.

Microfibers which were classified spectroscopically as cellulosic were further distinguished as anthropogenic cellulose (AC) fibers or cellulose fibers of unknown origin (UN) (i.e., possibly natural) (Athey et al., 2020). Cellulose morphology was confirmed when viewed at

Table 1
Locations of sampling sites, sediment depths and masses used for analyses, and total AP concentrations. Sites are numbered by longitude, west to east.

Sample Yr	Site #	Stn code	Lat (°N)	Long (°W)	Location	Water depth (m)	Sample mass (g)	Total APs (particles g^{-1} dw)	Sample depth (cm)
2015	1	CB2	71.4093	−157.4810	Beaufort Sea - Alaska	128	11.8	4.7	0–1
2014	2	GSC_9PCBC	70.6395	−139.0148	Beaufort Sea - Yukon	1502	15.7	1.4	0–5
2014	3	GSC_11BC	70.6303	−136.1890	Beaufort Sea - Mackenzie Delta	504	18.0	1.9	0–5
2014	4	GSC_1PCBC	72.1656	−130.8177	Beaufort Sea - East	961	23.6	0.7	0–5
2017	5	QMG-1	68.4925	−99.8980	Queen Maud Gulf	40	61.8	0.9	0–2
2017	6	3.4	71.4753	−91.9932	Gulf of Boothia	270	56.5	0.7	0–2
2017	7	304	74.2463	−91.5131	Northwest Passage West	315	58.8	1.0	0–2
2017	8	301	74.2777	−83.3194	Northwest Passage East	715	36.8	1.6	0–2
2015	9	155	72.4878	−78.7832	North Baffin Island	376	8.85	4.0	0–1
2017	10	101	76.3575	−77.5152	North Baffin Bay	378	52.0	0.8	0–2
2014	11	Kane2b	79.5152	−70.8470	Nares Strait	343	22.9	1.3	0–5
2017	12	8.1	69.4072	−64.7322	Mid-Baffin Island	1055	16.8	1.8	0–2
2015	13	177	67.4760	−63.7912	South Baffin Island	365	30.5	0.6	0–1
2014	14	105	73.2609	−57.8888	East Baffin Bay - Greenland	343	16.9	2.1	0–5

500–1000× magnification using the Royal Microscopical Society's Microscopy of Textile Microfibers Handbook (Greaves and Saville, 1995) and the guidelines of Zhu et al. (2019). AC fibers were identified based on (1) spectroscopic confirmation of cellulose (acknowledging that cellulose has a weak signal compared to that of synthetic materials (Lenz et al., 2015; Bart, 2006)), and one or both of (2) the signal of an additive, and/or (3) morphological evidence of anthropogenic alteration (e.g., extrusion). UN microfibers were those microfibers spectroscopically confirmed as cellulose, but lacking evidence of anthropogenic modification. Thus, UN microfibers included microfibers that (1) showed coloring that was deemed non-anthropogenic (e.g., grey, clear, yellow, green, brown, white, or black), (2) had no spectrometric signal of an associated chemical additive or polymer, and (3) showed no anthropogenically altered morphology. These UN microfibers therefore may or may not have had an anthropogenic source, thus they are not included in the reported confirmed AP concentrations below. All raw data (including numbers of UN) are reported in the SI. Concentrations are reported in MFs or MPs g^{-1} dry mass of sediment, and presented in Table S1 and in the figures in units of particles kg^{-1} dry mass of sediment, for ready comparison with literature values. Counts were arithmetically scaled from g to kg with no correction factors.

2.4. QA/QC

Throughout the collection and laboratory processing of samples, precautions were taken to minimize potential contamination: samples were covered at all times when not being processed, all materials were made of glass or stainless steel, unless otherwise stated. All materials used in sample processing were cleaned with soap and single-filtered RO water prior to and between uses.

Three spike and recovery trials were conducted prior to commencing sample processing to assess the reliability and appropriateness of sieving, and $CaCl_2$ density separation methods. Five different types and densities of microplastics and two types of microfibers were used in the trials: red opaque cellulose acetate beads (1.4 g/cm^3, 1000 μm; CAS-RED 1.0 mm), clear polystyrene beads (PS; 1.05 g/cm^3, 990 μm; PSS-1.05 0.99 ± 0.025 mm), clear polyethylene tetraphthalate fragments (PET; 1.38 g/cm^3; various sizes all <5 mm), two types of opaque polyethylene beads (PE; 1. Blue, 1.08 g/cm^3, 300–355 μm; BLPMS-1.08 300–355 μm; and 2. White, 1.15 g/cm^3, 300–355 μm; WPMS-1.15 300–355 μm), nylon fibers (1.15 g/cm^3, various sizes), and rayon fibers (1.5 g/cm^3, various sizes). Cellulose acetate beads, PS beads, and PE beads were sourced from Cospheric LLC (California, United States of America), while PET fragments, and nylon and rayon fibers were constructed in the lab from larger materials. The particles used as "spikes" had unique coloration and morphology, distinguishing them from any APs native to the lake sediment used. Approximately 30 g dw of sediment from a freshwater lake in the Greater Toronto Area was spiked with 10 particles of each type. The same sieving and $CaCl_2$ separation methods were then applied. Recoveries across each particle type in each of the three trials were between 80 and 100% and thus were considered satisfactory (Table S4).

Field, laboratory, and oven blanks were collected to account for potential contamination of samples during the collection and processing stages. Two field blanks were collected during the 2017 sampling trip. Field blanks were taken by placing ~30 g of anhydrous sodium sulphate in 250-ml glass jars and leaving them open on deck from when the sediment corer landed on deck until the sediment sample was collected. The field blanks were processed using the same protocol as for sediment samples. A RO water blank was processed concurrently to field blank processing. Each field blank was corrected for the RO water blank by colour-category. Three laboratory blanks were processed and analyzed concurrently with samples to account for potential of airborne contamination during processing. Laboratory blanks consisted of RO water in a glass beaker remaining open to air during the processing of a single sample. An additional oven blank was run as samples were drying to account for oven contamination. The colour-category particle values from the oven blank were then added to each lab blank to account for total possible contamination during processing. Median particle values for each colour-category were then calculated for the oven + lab blanks (Table S5). That value was rounded up and subtracted from the respective colour-category value from each sample as the lab blank correction factor. After processing lab blanks, it was determined that field blank numbers were very low (one black fiber total) and within the error of the lab blanks, and so field blanks were not included in the blank correction. Particles in all blanks consisted of 100% microfibers, thus only microfibers concentrations were blank corrected by subtracting based on colour.

A second correction factor ("cellulose correction") was applied following blank correction, using results of Raman spectroscopy and microfiber morphology. Cellulose correction was applied to subtract UN microfibers, i.e., cellulose microfibers of unknown origin (UN), from raw microfiber totals. The final AP concentrations reported here include only confirmed anthropogenic fibers. They do not include cellulose of unknown origin (UN); thus, using this cellulose correction, we provide a conservative estimate of total anthropogenic particle concentrations.

The proportion of UN microfibers in each non-anthropogenic colour category identified by Raman spectroscopy was applied to the total post-blank correction number of APs in the respective non-anthropogenic colour groups across all sites (See Table S6 for proportions). For example, based on spectroscopic results, 78% of all clear microfibers were identified as UN (a cellulose spectroscopic signature, but absence of any anthropogenic signal such as dye signature or evidence of morphological alteration). Therefore, 22% of all clear microfibers were confirmed as anthropogenic (presence of a chemical additive or synthetic polymer signature) via Raman spectroscopy. This proportion was then applied to the blank-corrected total count of clear microfibers to report the number of confirmed clear microfibers of anthropogenic origin in the whole dataset. However, microfibers identified as UN may still have an anthropogenic origin, as demonstrated by Athey et al. (2020), who reported UN microfibers in wash water from blue jeans laundering. As no white microfibers underwent Raman spectroscopy, Raman correction data for white microfibers were not available and therefore a correction factor was not applied to white microfibers. However only two white microfibers were found across all samples, and so lack of white microfiber correction factor does not significantly alter results.

Following the cellulose correction, numbers were rounded up to the nearest whole number for the final corrected values for each microfiber colour. These corrected AP and microfiber values reported in the Results and discussion section below therefore represent the *minimum number* of confirmed APs and microfibers in Canadian Arctic marine sediments. The Arctic marine sediment data presented here are a detailed examination of the aggregated data presented by Athey et al. (2020); the study of Athey et al. (2020) did not undertake the cellulose correction on the Arctic microfiber data because that study focussed on the release of anthropogenic cellulose from laundering of blue jeans. Therefore, corrected AP concentration data, corrected microfiber concentrations and proportions, as well as proportions of certain colored microfibers (those considered as "natural colors" - clear, grey, brown, green, and yellow) differ between the two studies (See SI for further details).

2.5. Statistical analyses

Correlation analyses were used to compare corrected total AP, microfiber, and microplastic concentrations with sediment sample mass, water depth at sampling point, latitude, and longitude. Analyses were run on post-corrected total concentrations for each of AP, microfiber, and microplastic at each site, not distinguishing chemical classes (i.e., synthetic, semi-synthetic, anthropogenic cellulose). Spatial trends of total APs, microfibers, and microplastics were mapped using ArcMap 10.6.1 with ocean currents.

3. Results and discussion

3.1. Prevalence of anthropogenic particles in Canadian Arctic sediments

The blank-corrected concentration, reported as mean (median) ± SD of total APs in sediment was 1.7 (1.3) ± 1.2 APs g^{-1} dw, ranging from 0.62 (13 - Stn 177) to 4.7 (1 - CB2) APs g^{-1} dw (Fig. 2). Microfibers comprised 82% of APs, followed by fragments (15%), while films, foams, spheres, and fiber bundles cumulatively represented 3% (Fig. 2). Due to a lack of standardization in sampling, processing, and reporting methods among AP studies, we compared our results to recent literature reports at the level of "orders of magnitude", given that differences in sampling equipment used, density separation methods, and scope of data reported can all affect total concentrations (Table S1).

The sediments from our study contained unprecedentedly high concentrations of APs when compared with studies that examined both microplastics and microfibers in sediments, including those reported from more southerly locations in Canada. Details for all comparable studies can be found in Table S1. For example, mean AP concentrations were 85 APs kg^{-1} dw, 220 APs kg^{-1} dw, 862 APs kg^{-1} dw, and 980 APs kg^{-1} dw for Lake Erie nearshore sediments (Dean et al., 2018), Ottawa River sediments (Vermaire et al., 2017), Lake Ontario and Lake Huron nearshore sediments (Athey et al., 2020), and Lake Ontario nearshore sediments (Ballent et al., 2016), respectively. Canadian Arctic mean AP concentrations (Fig. 2) were also higher in comparison to mean AP concentrations from global locations, including the Shanghai rivers (802 ± 594 APs kg^{-1} dw; Peng et al., 2018) and estuaries (121 ± 94 APs kg^{-1} dw; Peng et al., 2017), the North Yellow Sea (37 ± 43 APs kg^{-1} dw; Zhu et al., 2018), Belgian harbours (167 ± 92 APs kg^{-1} dw; Claessens et al., 2011), the Northeast Atlantic Ocean (421 APs kg^{-1} dw; Maes et al., 2017), and the North Pacific Ocean to Chukchi Sea (22.9 APs kg^{-1} dw; Mu et al., 2019). Mean concentrations of APs in Canadian Arctic sediments were similar to mean concentrations found in sediments from highly populated urban Dutch Canals (2071 ± 4146 APs kg^{-1} dw; Leslie et al., 2017) and the Dutch North Sea coast (1400–4900 APs kg^{-1} dw; Leslie et al., 2017), as well as suburban shallow lakes of the Greater Toronto Area (2834 ± 1135 APs kg^{-1} dw; Athey et al., 2020). However, we emphasize the problematic nature of direct comparisons between studies, which remain difficult due to different size fractions considered by each study, different laboratory methodologies and approaches to

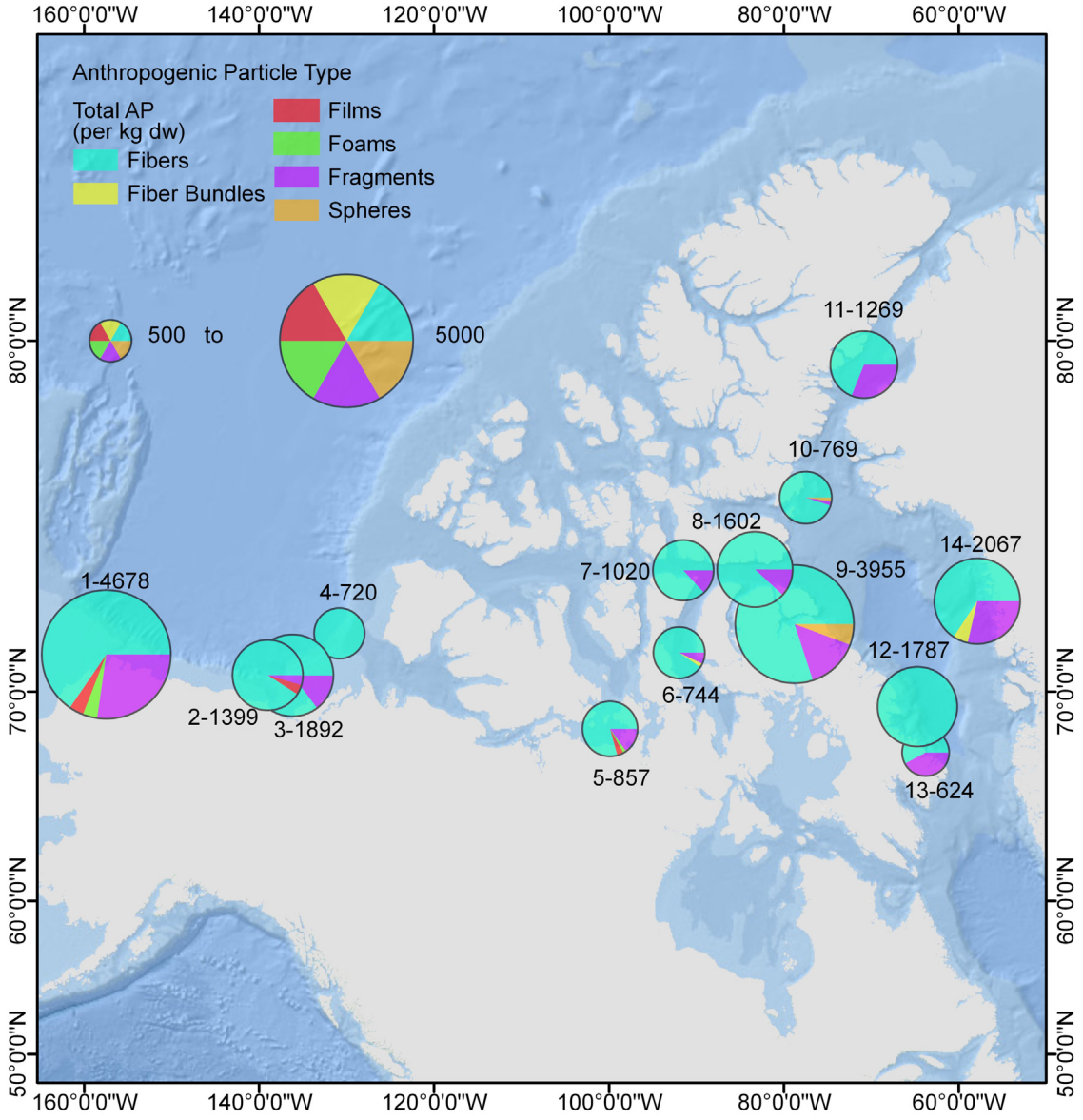

Fig. 2. Total concentrations of anthropogenic particles (APs) at each site, and proportions of each AP type. The size of each pie chart represents the total number of APs per kg dried sediment from each site. Numbers adjacent to each pie chart indicate the site number (Table 1), followed by the total number of APs kg^{-1} dw (Site # - AP concentration).

QA/QC, and the fact that many studies may either under-report microfiber concentrations, particularly those of cellulose microfibers, or choose not to report microfiber concentrations entirely.

Sediments from 6 of the 14 stations presented here (Sites 5–8, 10, and 12) were also analyzed by Huntington et al. (2020) in a comparative analysis of microplastic and microfiber concentrations in sediments, surface water and zooplankton. Samples presented here and those of Huntington et al. (2020) were collected from the same six sites during the same year (2017), however it is unknown if they are subsamples from the same core, or were taken from different cores. Lab handling and enumeration methods for anthropogenic particle extraction from sediments differed, including density separation methods and minimum sieve size. Three out of the six common sites (6, 7, and 12) had AP concentrations within the same order of magnitude while larger differences in concentrations were observed at the remaining three sites (Table 1, Table S1). These differences can be attributed to differences in the methods used to handle the samples, and they support the widespread call for the harmonization of methods so that results may be compared between studies.

We also note that sample mass analyzed (g) was negatively correlated with each of total AP, microfiber, and microplastic concentration (Fig. S3). Sample masses was set by availability of sediment, and it varied by a factor of 7 (9–62 g). The low number of samples in this study ($n = 14$) prevented us from determining whether this correlation was coincidental or methodological and are a limitation of the data. We note that future studies should consider the effect of sample mass and AP occurrence. Greater sample masses and lower variation in mass between samples, along with analysis of replicates, would allow for greater statistical power and representativeness (Prata et al., 2019; Cowger et al., 2020).

No trend was observed according to sampling year, but samples did vary in terms of sediment depths sampled. Sediment samples were taken from three different surface depths in the box corers (0–1 cm; 0–2 cm; 0–5 cm; Table 1); AP concentration was not significantly different between sites in each of these groups (ANOVA $F = 3.19$; $df = 2$; $p = 0.08$) but the highest concentration was recorded in the samples spanning the top 0–1 cm. This effect could add inter-site variability in concentrations that is independent of AP deposition/accumulation, and it may relate to variation in sedimentation rate. However, since sediment accumulation rates are so poorly known in the Canadian Arctic, are likely highly variable, and not available at the sampling stations, this effect is difficult to assess. Bailey et al. (2013) estimate 0.04 cm/yr in Baffin Bay, which would mean that the entire post-1950 CE era of dramatic growth in plastic production (Geyer et al., 2017) is included within the top 3 cm.

Our results point to Arctic marine sediments as a zone of high deposition for APs. This is consistent with the findings of other studies documenting the widespread accumulation of APs in the Arctic where concentrations can far exceed those of source areas (e.g. Lusher et al., 2015; Bergmann et al., 2017; Cózar et al., 2017; Kanhai et al., 2018a; Huntington et al., 2020), indicating that circumpolar sediments are a geographic sink for APs. This is consistent with previous studies that have shown Arctic sediments act as a sink for APs and other numerous contaminants, including legacy contaminants such as PCBs and organochlorine pesticides (Macdonald et al., 2000; AMAP, 2017), as well as contemporary contaminants such as organophosphate esters (Ma et al., 2017; Sühring et al., 2021) and siloxanes (Panagopoulos-Abrahamsson et al., 2020).

3.2. Physical and chemical characterization of microfibers

Fibers, which were the dominant form of APs, had a mean (median) concentration of 1.3 (1.1) ± 0.99 fibers g^{-1} dw, ranging from 0.44 (Site 13 – Stn 177) to 3.2 (Site 9 – Stn 155) fibers g^{-1} dw (Figs. 2, 3). The maximum concentration at Site 9 was closely followed by Site 1 – CB2, with 3.1 fibers g^{-1} dw. Whereas fibers were found at all 14 sites, fiber bundles were found at 2 sites: Site 6 (Stn 3.4; 0.020 fiber bundles g^{-1} dw), and Site 14 (Stn 105; 0.12 fiber bundles g^{-1} dw). Microfibers (single fibers and fiber bundles, combined) ranged from 58 to 100% of APs at a single site, and represented 100% of APs found at 2 sites: Site 4 (GSC_1PCBC) and Site 12 (Stn 8.1), but only 58% of APs at Site 13 (Stn 177) (Fig. 2). Blue microfibers comprised 59% of all microfibers, followed by clear at 11% (Fig. 3). Corresponding site numbers, station codes, and geographic location names can be found in Table 1.

As with the mean concentrations of total APs, mean microfiber concentrations in this study were higher than mean concentrations found in sediments both in southern Canadian freshwater systems, including Lake Erie nearshore sediments (57 microfibers kg^{-1} dw; Dean et al., 2018), and Lake Ontario + Lake Huron nearshore sediments (780 ± 440 microfibers kg^{-1} dw; Athey et al., 2020), and international locations closer to sources, including the Northeast Atlantic Ocean (163 microfibers kg^{-1} dw, range = 99 to 301 microfibers kg^{-1} dw; Maes et al., 2017), the Belgian Coast (66.3 ± 28.6 microfibers kg^{-1} dw; Claessens et al., 2011), and Shanghai estuaries (112 microfibers kg^{-1} dw; Peng et al., 2017). Our results were of the same order of magnitude as microfiber abundances in shallow suburban lakes in the Greater Toronto Area (2490 ± 1260 microfibers kg^{-1} dw; Athey et al., 2020). A prevalence in AP assemblages of microfibers has also been shown elsewhere in freshwater (e.g. Vermaire et al., 2017; Athey et al., 2020) and marine sediments (e.g. Nor and Obbard, 2014; Woodall et al., 2014; Reed et al., 2018; Mu et al., 2019; Huntington et al., 2020) (Table S1). Lusher et al. (2015) suggested that the dominance of microfibers in Arctic samples nearby Europe was attributable to local sources, such as commercial and recreational activities, and wastewater. However, Huntington et al. (2020) found no correlation between AP concentrations and local population densities in the Eastern Canadian Arctic, suggesting microfibers in that region are likely subject to long-range transport via ocean or air. Athey et al. (2020) reached a similar conclusion that long-range transport was the most likely reason for the prevalence of indigo denim fibers at these sites. This difference in source attribution between the near-Europe vs Canadian archipelago may be due to the sparser population densities in the Canadian Arctic than the European Arctic. Finally, the dominance of microfibers in this study and others is a clear indication that many microplastic studies which exclude microfibers severely underestimated total concentrations of APs (Table S1).

Of the 146 microfibers which underwent Raman spectroscopy, 57% were cellulosic, 11% were synthetic (either polyester, polyacrylonitrile, or polyurethane), and 32% emitted a dye signature only (Fig. 4). Dyes observed on microfibers included azo dyes (Amido Black 10B and Sirius Light Green), reactive dyes (Levafix blue), and vat dyes (Indigo). The two most common dyes were Amido Black 10B and Indigo, present on 25% and 21% of all microfibers analyzed, respectively. Particular dyes are used in conjunction with certain types of textiles during production (Zhu et al., 2019). The two common dyes encountered in the samples are most often used to dye cotton. Indigo is used almost exclusively to dye cotton in denim textiles (Raina et al., 2015) and Amido Black 10B is an azo dye which is most often used on cotton, and has minor uses for silk, wool, viscose, and some synthetics (Garg et al., 2015). Azo dyes have been found to be toxic, causing damage to DNA (da Cruz Brambilla et al., 2019), destroying lung tissue, and irritating eyes and skin (Mittal et al., 2013). A standard approach of analyzing 10% of each colour category by Raman spectroscopy was used here (Grbić et al., 2020), resulting in chemical characterization of 22% of all particles and fibers of confirmed anthropogenic origin (APs). We acknowledge that increasing the sample size for Raman chemical characterization would provide additional detail on the plastic and fiber types.

Our findings here indicate the high abundance and widespread prevalence of anthropogenic cellulose (AC) fibers in Arctic sediments. AC fibers were the most abundant and also prevalent form of microfiber and AP, with AC fibers found across all 14 sites studied. Additionally, of the 47 microfibers which emitted only a dye signature, 79% were

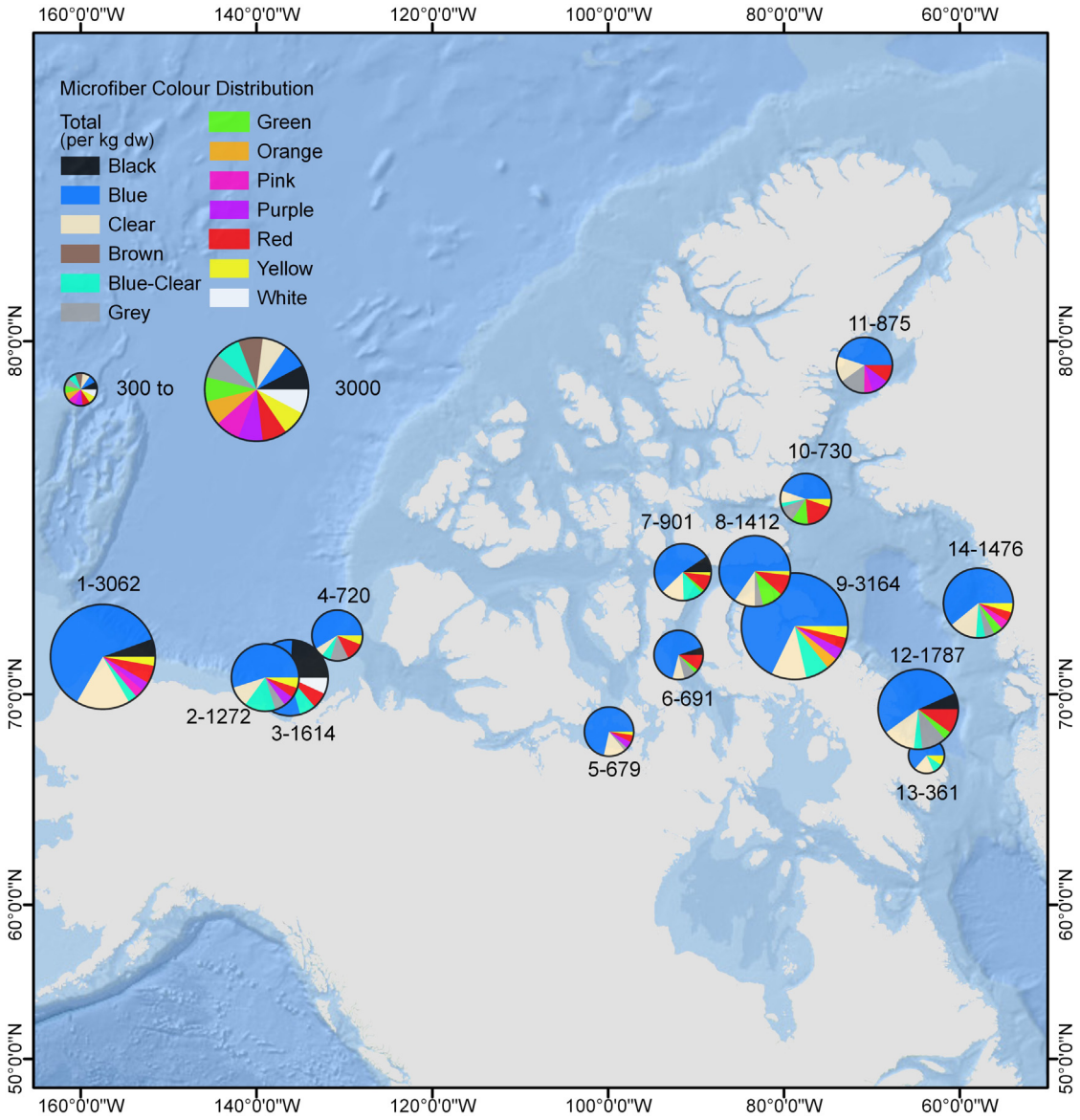

Fig. 3. Total microfiber (MF) concentration at each site, and proportions of each microfiber colour. The size of each pie chart represents the total number of MFs per kg dried sediment from each site. Numbers adjacent to each pie chart indicate the site number (Table 1), followed by the total number of MFs kg^{-1} dw (Site # - MF concentration).

visually identified as cellulose according to morphology. Of the 146 microfibers analyzed by Raman spectroscopy, 51% (75 fibers) were classified as AC fibers and 31% (45 fibers) were UN fibers. The UN fibers were not included in the total reported concentrations as they could be of either natural or anthropogenic origin. The flux of organic matter to marine sediments through the biological pump is a key natural process in global carbon cycling. Cellulose is a complex carbohydrate making up the structural tissues of green plants and is a major component of terrestrial and near-shore vegetation (e.g., Trevathan-Tackett et al., 2017). Cellulose is moderately resistant to microbial decomposition; thus naturally-derived cellulose from terrestrial and near-shore settings is expected to be deposited in marine sediments following transport through watersheds and coastal zones (Arndt et al., 2013), and it is likely to be present in a surface sediment sample. Thus, it is critical for microfiber studies to distinguish between cellulose naturally occurring as a result of the biological pump, and cellulose microfibers originating from textiles, as we have done here.

Our inclusion of AC fibers likely contributes to our finding of exceptionally high concentrations of APs in Canadian Arctic sediments in comparison to other studies (Table S1). Studies which have previously focused only on synthetic microfibers are likely to underestimate the spatial extent of APs and their abundances. Here, only 64% (9 of 14) of Canadian Arctic sites contained synthetic microfibers. Synthetic microfibers were not found at Sites 9, 10, 12–14, all in the Eastern Canadian Arctic. AC fibers are rarely reported, yet the few studies which have reported cellulosic microfibers also find them to dominate microfiber types in environmental samples (e.g. Kanhai et al., 2018b; Sanchez-Vidal et al., 2018; Stanton et al., 2019). However, these studies do not differentiate possible AC from UN cellulose microfibers, and thus may overestimate AC concentrations. AC fibers thus require future consideration using emerging analytical methods (e.g., Karakolis et al., 2019; Zhu et al., 2019).

The main concern associated with the widespread contamination and persistence of AC fibers in the environment are their abundance, as well as their high chemical additive burden. For example, cotton (the most commonly produced cellulosic textile fiber) contains numerous additives. These additives may include organophosphate esters (OPEs), bisphenols, as well as synthetic dyes (Abou-Okeil et al., 2013; Zhu et al., 2020). Additionally, cotton textiles have been shown to accumulate contaminants, such as brominated and OPE flame retardants

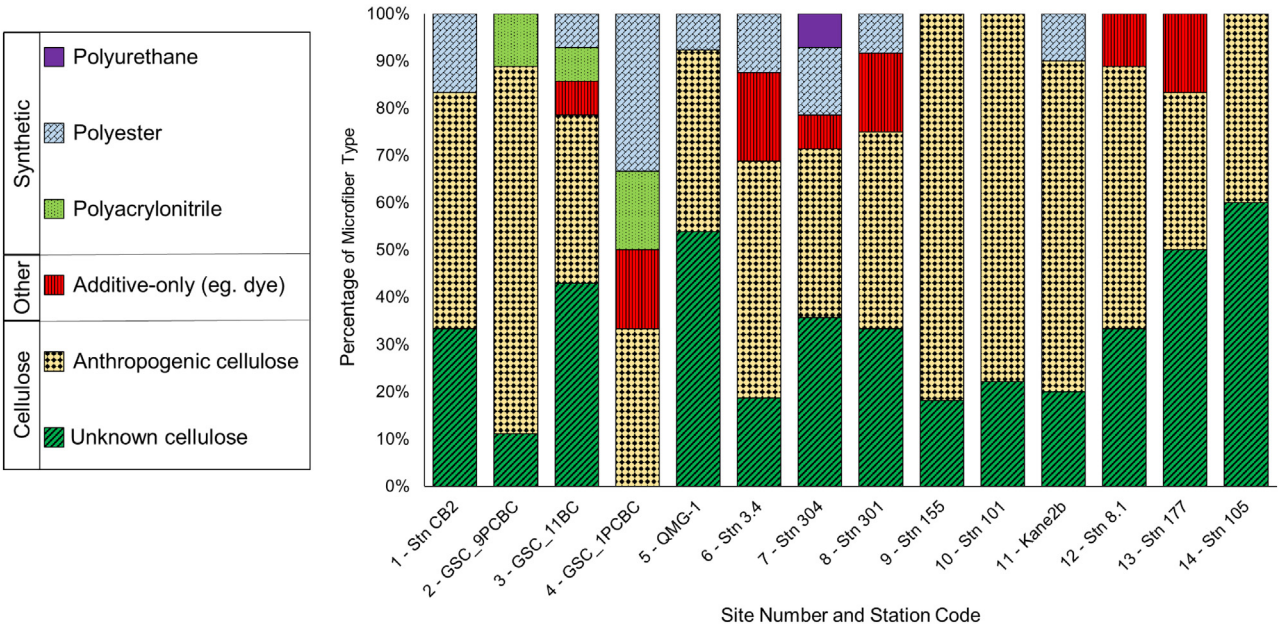

Fig. 4. Composition of microfibers (MFs) as determined by Raman spectroscopy for $n = 146$ MFs, a subset (21%) of all MFs recovered from samples. Site are ordered by longitude, west to east. Raw data are provided in Table S9a.

and phthalates, from the ambient environment (Saini et al., 2017; Li et al., 2019). These chemical additives may influence the persistence of these fibers in the marine environment. While several studies have recorded the presence of microfibers in foraging Arctic seabirds (Amelineau et al., 2016; Provencher et al., 2018), and in a key fish species in the Eurasian Arctic, polar cod (*Boreogadus saida*) (Kühn et al., 2018; Morgana et al., 2018), exposure to and effects from AC microfibers for biota are currently unknown. Our study confirms that these fibers are accumulating in high concentrations in Arctic marine sediments.

It is suspected that the major source of microfibers is from shedding of fibers from clothing during use and laundering (De Falco et al., 2020), and most studies to date have examined only the release of synthetic microfibers from such sources (e.g. Browne et al., 2011; Hartline et al., 2016; Gago et al., 2018). Microfibers may also originate from the degradation of fish nets and lines, shedding of furniture and carpets, artificial turf, and application of microfiber-laden wastewater treatment plant sludge to agricultural fields (Dris et al., 2016; Magnusson et al., 2016).

3.3. Physical and chemical characterization of microplastics

Microplastics (fragments, films, foams, and spheres) had a mean (median) concentration of 0.33 (0.22) ± 0.43 microplastics g^{-1} dw; ranging from not detected at Site 12 (Stn 8.1) and Site 4 (GSC_1PCBC), to 1.6 microplastics g^{-1} dw at Site 1 (CB2) (Fig. 5). Microplastics constituted 0 to 42% of total APs at a single site (Fig. 2). Fragments were found at 12 of the 14 sites at a mean concentration of 0.22 ± 0.44 fragments g^{-1} dw. Films were found at 3 of the 14 sites: Site 2 (GSC_9PCBC), Site 1 (CB2), and Site 5 (QMG-1), with a maximum concentration of 0.22 films g^{-1} dw. Foams were found at 2 of the 14 sites: Site 1 (CB2) and Site 5 (QMG-1), with a maximum concentration of 0.22 foams g^{-1} dw. Finally, spheres were also found at 2 of the 14 sites: Site 10 (Stn 101) and Site 9 (Stn 155), at a maximum concentration of 0.23 spheres g^{-1} dw. Clear was the most common colour at 44% of all microplastics (Fig. 5).

Mean concentrations of microplastics within the present study were similar to mean concentrations of microplastics found in sediments from southern Canadian locations, including in Lake Ontario (550 microplastics kg^{-1} dw; Ballent et al., 2016), as well as international locations including the English channel (350 microplastics kg^{-1} dw; Maes et al., 2017), and the Dutch coast (123 microplastics kg^{-1} dw; Maes et al., 2017). Mean MP concentrations were also similar to those found in the Eastern Canadian Arctic (400 microplastics kg^{-1} dw; Huntington et al., 2020). However, mean microplastic concentrations in this study were higher than those found in sediments from southern Canada in the Ottawa River (<11 microplastics kg^{-1} dw; Vermaire et al., 2017), and international locations of the Bering Strait/Chukchi Sea (8 microplastics kg^{-1} dw; Mu et al., 2019), the North Yellow Sea (25 microplastics kg^{-1} dw; Zhu et al., 2018), the Belgian Coast (30 microplastics kg^{-1} dw; Claessens et al., 2011), and Shanghai estuaries (10 microplastics kg^{-1} dw; Peng et al., 2017). However, size fractions examined in previous studies varied widely (Table S1), making direct comparisons difficult.

Of the 37 non-fibrous particles that underwent Raman spectroscopy, 70% were synthetic particles (including synthetic microplastics and tire wear particles), followed by cellulosic particles (22%), and those which only emitted a dye signal (8%; Vine Black and Irgdlith Blue). Forty-two percent of synthetic particles were composed of polyvinyl chloride (PVC), which dominated the synthetic polymers. Other synthetic polymers found included polyacrylamide, polystyrene, polyurethane, and polyethylene. Additionally, 23% of all synthetic particles (16% of all non-fibrous particles) were characterized as suspected tire wear particles, based on texture and carbon black Raman spectra (Kreider et al., 2010; Lenz et al., 2015). Of the cellulosic particles, 88% (7 particles) were unknown origin cellulose and 12% (1 particle) were anthropogenic cellulose, based on the presence of an additive. This equated to 19% and 3% of all microplastics being of unknown origin and anthropogenic cellulose, respectively (Fig. 6). Foams were identified as either PVC or unknown origin cellulose, and films were polyacrylamide or PVC. Of the three spheres found across all samples, none underwent Raman spectroscopy.

3.4. Geographic distribution of anthropogenic particle accumulation

Total APs, microfibers, and microplastics were not correlated to latitude, longitude, or bottom depth (Fig. S3). Despite low correlations between concentrations and geospatial variables, APs, microfibers and microplastics tended to have high concentrations in two regions: the Beaufort Sea in the Western Arctic and North Baffin Island/Baffin Bay in the Eastern Arctic (Fig. 2). While a comprehensive assessment of geographical trends of AP, microfiber, and microplastics concentrations was not possible due to the low number of sites ($n = 14$), the complexity of

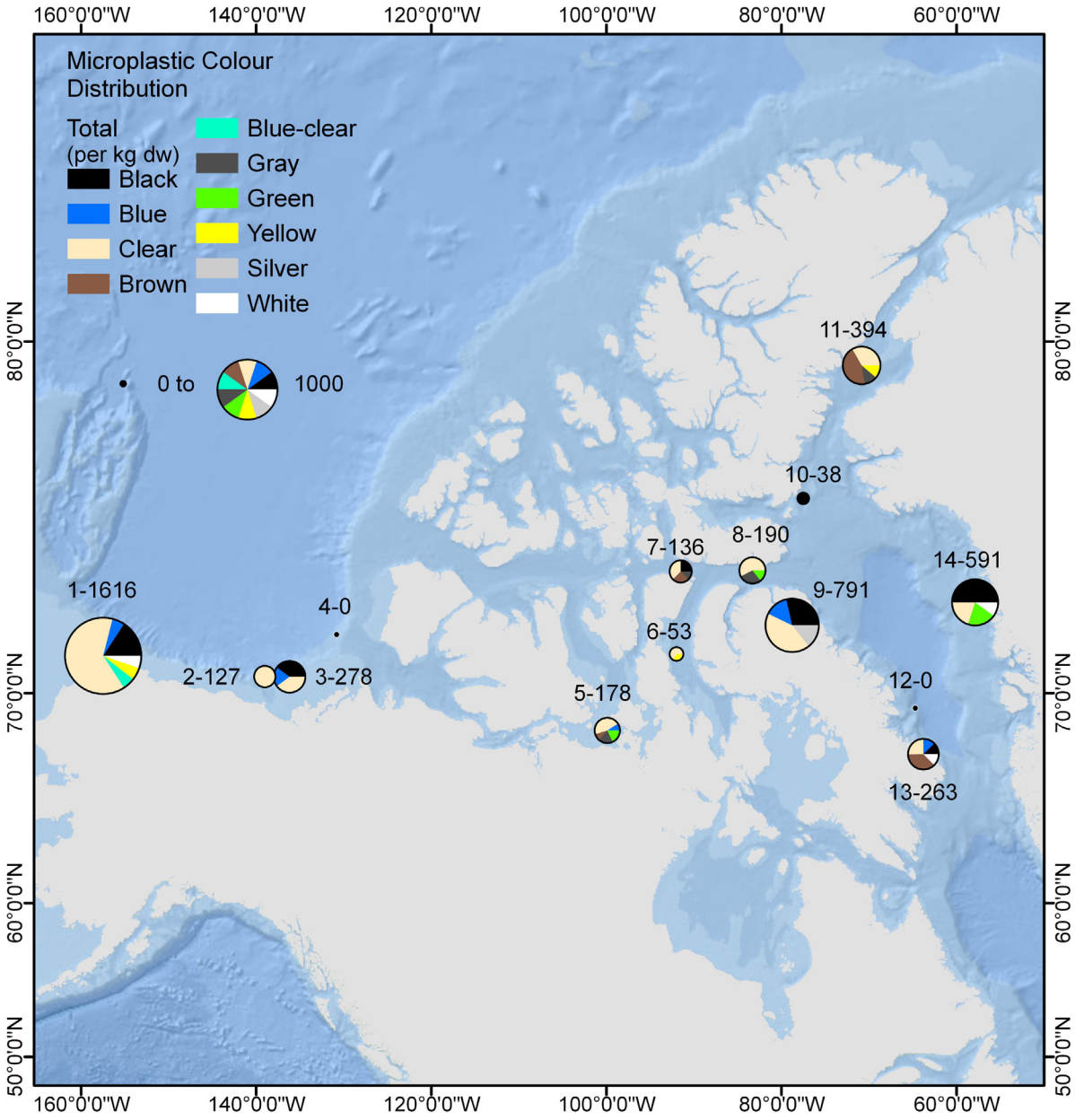

Fig. 5. Total microplastic (MP) concentrations at each site, and proportions of each microplastic colour. The size of each pie chart represents the total number of MPs per kg dried sediment from each site. Numbers adjacent to each pie chart indicate the site number (Table 1), followed by the total number of MPs kg^{-1} dw (Site # - MP concentration).

water movements in the Canadian Arctic, and unknown rates of sediment accretion, the finding of clusters of sites or single locations with high concentrations may help inform future research.

Higher concentrations in the Beaufort Sea (Sites 1–4) have two possible explanations. First, the nearby Mackenzie River may transport APs deposited atmospherically and sourced from within the drainage basin (e.g. Hurley et al., 2018). Moreover, major rivers have been found to be an important transport vector for siloxanes and OPEs to the Canadian Arctic (Panagopoulos-Abrahamsson et al., 2020; Sühring et al., 2021) The Mackenzie River contributes ~60% of the total river discharge into the Arctic from Canada, with a large drainage basin area (1.8 million km^2) encompassing a region with a population of ~400,000, which is heavily industrialized (e.g. Alberta oil sands regions) (Mackenzie River Basin Board, 2003). The Mackenzie River discharge zone is a hotspot for deposition of organic carbon across the Canadian Arctic. Previous studies have found a significant positive relationship between organic carbon and AP concentrations, perhaps due to similar transport and deposition processes (Strand et al., 2013; Maes et al., 2017). Second, APs along with sediment and organic matter, could be transported by Pacific Ocean currents from the south, particularly Asia, through the Bering Strait into the Beaufort Sea, where water velocity slows due to the presence of sills on the edge of the Canadian archipelago, causing deposition of particle load (Macdonald and Bewers, 1996; Zarfl and Matthies, 2010; Sigler et al., 2017; Mu et al., 2019; Wichmann et al., 2019).

High concentrations of APs occurred in two locations within Baffin Bay (Sites 12 and 14). We postulate that these could originate in part from oceanic and/or atmospheric transport. The counter-clockwise movement of ocean currents from the Labrador Sea (Fig. 1) could transport APs from the Atlantic Ocean into Baffin Bay, resulting in an AP "sink" in the Eastern Canadian Arctic. Similarly, Benskin et al. (2012) and Brown et al. (2018) concluded that Atlantic surface currents play a dominant role in bringing PFAAs and PFOA compounds to the Eastern Canadian Arctic from southerly locations. Additionally, legacy contaminants that have accumulated in the Central Arctic region over the past few decades are transported into Baffin Bay via ocean currents (Brown et al., 2018). Atmospheric deposition has been documented elsewhere

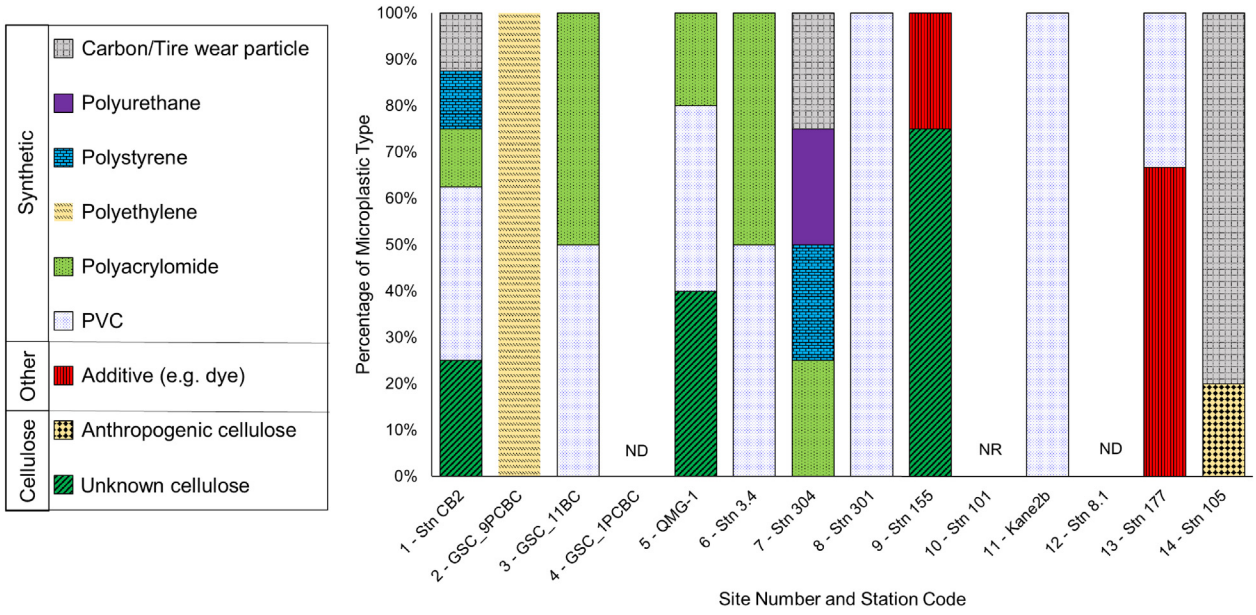

Fig. 6. Composition of non-fiber microplastics (MPs) as determined by Raman spectroscopy for $n = 37$ MPs, a subset (34%) of all MPs recovered from samples. Sites are ordered by longitude, west to east. ND indicates sites at which MPs were not found. NR indicates that MPs found at this site did not undergo Raman spectroscopy. Raw data are provided in Table S9b. The term microplastics here includes all non-fibrous particles (<5 mm) of anthropogenic origin including plastics, tire wear particles and cellulosic foams or fragments.

as an important vehicle for long-distance transport of APs (Evangeliou et al., 2020) and may also be an important source of APs to this region compounding the oceanic transport from the Atlantic Ocean into Baffin Bay via the Labrador Sea. Rates of atmospheric deposition, however, have not been measured for the Canadian Arctic.

Local sources have been noted to contribute to organic pollutant contamination across the Canadian Arctic, and local sources may also be important for APs in the Arctic. Sewage lagoons have been suggested to contribute to the accumulation in the European Arctic of APs and other anthropogenic contaminants (Lusher et al., 2015; Gewurtz et al., 2020). The Beaufort Sea-Alaska site (Site 1) is located just offshore of Utqiagvik (Barrow), Alaska, with a relatively high population of ~4400 in 2018 (United States Census Bureau, 2018). High concentrations of microplastics in the Beaufort Sea, dominated by high-density polymers (PVC, cellulose, polyacrylamide) and tire-wear particles, suggest some potential contribution from local sources (Lusher et al., 2015).

In the Eastern Canadian Arctic, the sampling site with the highest microfiber concentration was located by North Baffin Island (Site 9), in close proximity to Bylot Island. Bylot Island contains one of the largest seabird colonies in the Arctic (Provencher et al., 2018). Arctic seabirds have been found to be concentrators of APs as they can transport APs via ingestion from a 50 km-wide foraging range followed by expulsion in guano or use as nesting material at colonial nesting sites (Provencher et al., 2015; Provencher et al., 2018). However, this site also had the lowest sample mass (~9 g), which may have affected the representativeness of the reported concentration (Fig. S3).

Local anthropogenic activities, such as shipping, may also increase the load of APs to a region (Wang et al., 2018). The Eastern Arctic is a common Arctic shipping route for commercial and passenger cruises (Fig. 1), and one of the most frequented shipping ports in the Canadian Arctic, Milne Inlet, is located on northern Baffin Island in close proximity to Site 9 (Chan et al., 2013). Other sites with high AP abundances located in East Baffin Bay, along the Northwest Passage and adjacent to Baffin Island, tend to be common stops for Arctic cruises. Additional data are needed to evaluate the importance of local sources in the Canadian Arctic.

Climate change may accelerate AP accumulation in the Arctic. High concentrations of APs in East Baffin Bay adjacent to the west coast of Greenland may be linked to the recent warming of subsurface waters along the coast (Holland et al., 2008). This has promoted recent accelerated melting of the Greenland ice sheet, increased runoff of surface meltwater, and increased ice discharge into the surrounding ocean (Hanna et al., 2013; King et al., 2018) thus causing a release of APs previously trapped within the ice (Obbard et al., 2014; Ambrosini et al., 2019), and accelerated deposition of APs into Baffin Bay. The extent of ice degradation in the Canadian Arctic is accelerating every year, exemplified by the recent collapse of the Milne Ice Shelf off of Ellesmere Island in the Northern Canadian Arctic on August 7th 2020 (Environment and Climate Change Canada, Canadian Ice Service). Multi-year ice has been implicated as both a sink and transport mechanism of APs within the Arctic (Bergmann et al., 2017; Peeken et al., 2018). The extent and duration of multi-year ice is declining due to climate change, which could affect the distribution and concentrations of APs in the Arctic; as multi-year ice melts and is not reformed, sea ice then becomes a secondary source of APs back into the Arctic environment (Obbard et al., 2014; Peeken et al., 2018). Oceanic circulation patterns may lead to the transport of recently released APs from melting sea ice in the central Arctic Ocean into Baffin Bay.

4. Conclusions

This study revealed the prevalence and extent of APs (microfibers and microplastics) across the remote Canadian Arctic and highlights the global nature of microfiber and microplastic pollution. While our study did not explicitly evaluate sources of APs, the results showing elevated concentrations of APs suggest that Arctic sediments are a sink for these contaminants. Sediment samples were dominated by AC fibers, while synthetic or plastic fibers were low in concentration. The results further corroborate emerging indications for the prevalence of anthropogenically modified cellulose fibers. The high concentrations reported here throughout the Arctic, coupled with evidence of atmospheric, oceanic and riverine transport of APs (e.g., Evangeliou et al., 2020), suggest contributions from long-range transport as well as from local communities and ship traffic. The inclusion of AC fibers in total AP concentrations resulted in higher reported values compared to previous reports, suggesting that past studies underreport microfiber pollution in sediments. Sites by Northern Baffin Island, Baffin Bay and the Beaufort Sea emerged as areas of higher AP concentrations, likely due to a combination of long-range transport and local sources. However low sample masses, low sample numbers ($n = 14$) and lack of replication limit the strength of this conclusion.

Future research should investigate sources and transport mechanisms of APs to the Arctic, such as communities, ship traffic and bird colonies. Additionally, as the Arctic is a region experiencing rapid climate change, research on the effects of change on AP accumulation in a region already characterized with high AP concentrations should also be prioritized. Lastly, high AP concentrations observed in the Beaufort Sea and Canadian Arctic in general, may have the potential to impact local organisms, and traditional food sources of Indigenous peoples. Yet, the effects of high concentrations of APs on Arctic wildlife, as well as high concentrations of anthropogenic cellulose fibers on Arctic aquatic organisms remain unknown and require further study.

CRediT authorship contribution statement

Jennifer K. Adams: Investigation, Formal analysis, Data curation, Funding acquisition, Writing – original draft. **Bethany Y. Dean:** Formal analysis, Data curation, Writing – review & editing, Visualization. **Samantha N. Athey:** Investigation, Formal analysis, Data curation, Writing – review & editing. **Liisa M. Jantunen:** Conceptualization, Resources, Funding acquisition, Writing – review & editing. **Sarah Bernstein:** Resources. **Gary Stern:** Resources. **Miriam L. Diamond:** Conceptualization, Writing – review & editing, Funding acquisition, Supervision. **Sarah A. Finkelstein:** Data curation, Writing – review & editing, Funding acquisition, Supervision.

Declaration of competing interest

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

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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.2021.147155.

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