Marine environment microfiber contamination: global patterns and the diversity of microparticle origins

A.P.W., Barrows,^{1,2*} S.E., Cathey,^{1,3} C.W., Petersen²

1. Adventure Scientists, PO Box 1834, Bozeman, MT 59771 USA.

2. College of the Atlantic, Department of Biology 105 Eden Street, Bar Harbor, ME 04609, USA.

3. Virginia Polytechnic Institute and State University, Department of Biological Sciences
926 West Campus Drive Blacksburg, VA 24061

*Corresponding author email: abarrows@coa.edu

Abstract

Microplastic and microfiber pollution has been documented in all major ocean basins. Microfibers are one of the most common microparticle pollutants along shorelines. Over 9 million tons of fibers are produced annually; 60% are synthetic and ~25% are non-synthetic. Non-synthetic and semi-synthetic microfibers are infrequently documented and not typically included in marine environment impact analyses, resulting in underestimation of a potentially pervasive and harmful pollutant. We present the most extensive worldwide microparticle distribution dataset using 1-liter grab samples (n = 1393). Our citizen scientist driven study shows a global microparticle average of 11.8 ± 24.0 particles L⁻¹ (mean $\pm$ SD), approximately three orders of magnitude higher than global model predictions. Open ocean samples showed consistently higher densities than coastal samples, with the highest concentrations found in the polar oceans (n = 51), confirming previous empirical and theoretical studies. Particles were predominantly microfibers (91%) and 0.1 - 1.5 mm in length (77%), a smaller size than those captured in the majority of surface studies. Using μ FT-IR we determined the material types of 113 pieces; 57% were classified as synthetic, 12% as semi-synthetic, and 31% as non-synthetic. Samples were taken globally, including from coastal environments and understudied ocean regions. Some of these sites are emerging as areas of concentrated floating plastic and anthropogenic debris, influenced by distant waste mismanagement and/or deposition of airborne particles. Incorporation of smaller-sized

microfibers in oceanographic models, which has been lacking, will help us to better understand the movement and transformation of synthetic, semi-synthetic and non-synthetic microparticles in regional seas and ocean basins.

Keywords: *microfiber; microplastic; marine pollution; microparticle; citizen science*

Capsule: This study increases understanding of global microplastic distribution in understudied regions and highlights the prevalence of synthetic and non-synthetic microfiber pollution.

Introduction

Plastic is a major pollutant throughout the world. It is one of the most prolific materials manufactured globally, with over 322 million tons produced annually with the majority of the production going to single-use packaging (PlasticsEurope, 2016). The disposable nature of this plastic type generates a high volume of packaging that continuously enters the waste stream. Plastics are cheap, lightweight, and durable—characteristics that have made it an ever more attractive packaging material and led to its high volume in solid waste streams. Plastics are now a common and persistent pollutant. Most waste management infrastructure worldwide does not match disposal needs, with an estimated 4.8 to 12.7 million tons of coastal plastic waste entering the ocean each year (Jambeck et al., 2015). Rivers are also a global vector for plastic and are estimated to transport between 1.15 and 2.41 million tons of plastic waste into the ocean annually (Lebreton et al., 2017). Consequently, between 5.95 and 15.11 million tons of plastic enter the ocean via coastal land and inland rivers (Lebreton et al., 2017).

Plastic waste estimates are often based on mesoplastic (5 mm - 2.5 cm) and macroplastic (2.5 cm - 1 m) lengths. Larger plastic has long been the focus of public concern, mostly due to its visibility and documented negative interaction with animals (Gall and Thompson, 2015; Zettler et al., 2017). However, plastics also enter the ocean as microplastic (particles less than < 5 mm in size) through storm drains, run-off, wastewater treatment plant outfall pipes, tire wear, and atmospheric deposition, among other sources (Arthur et al., 2009; Auta et al., 2017; Browne et al., 2011; Dris et al., 2017; Galgani et al., 2015; Verschoor, 2014). Microplastics are categorized as either primary, meaning manufactured to be less than 5 mm, or secondary, which are plastics that are less than 5 mm as the result of the fragmentation of a larger plastic piece (Andrady, 2011;

69 Cole et al., 2011). The relative significance of the two microplastic types are not well studied
70 (Boucher and Friot, 2017), but the majority of particles in surface water appear to be microfibers,
71 a threadlike particle with a length between 100 μm and 5 mm and a width of approximately 1.5
72 orders of magnitude shorter (Barrows et al., 2017), suggesting secondary microplastics are
73 predominant (Browne et al., 2011; Carr, 2017; Mason et al., 2016).

74

75 Of the 9 million tons of fibers produced globally in 2016, cotton accounted for 30%, wool, silk
76 and other natural fibers accounted for another 10%, and the remainder was synthetic (Carr, 2017).
77 Non-synthetic and semi-synthetic (e.g. rayon) fibers have infrequently been reported in surface
78 water studies. The few cases where they have been noted have been predominately in ingestion
79 studies (Lusher et al., 2013; Remy et al., 2015; Rochman et al., 2015; Zhao et al., 2016). The
80 majority of non-synthetic fiber textiles are treated with a similar cocktail of dyes and chemicals as
81 synthetic textiles and can accumulate chemicals from the ambient water (O'Neill et al., 1999;
82 Remy et al., 2015). These non-synthetic and semi-synthetic microfibers and their additives or
83 dyes may interact negatively with biota in aquatic environments similar to plastic microfibers, but
84 ingestion, chemical leaching and degradation rates in marine environments is poorly understood
85 (Remy et al., 2015). It is possible that, like 'biodegradable' plastic, non-synthetic microfibers
86 may not break down as readily as expected in the open ocean environment (Bagheri et al., 2017).

87

88 The majority of microplastic field sampling uses a trawl net. This allows for a large volume of
89 water to be sampled, but will miss many of the particles that can pass through the most commonly
90 used 333 μm mesh. This includes an unknown proportion of microfibers which can be many
91 millimeters long, but typically have a diameter smaller than most mesh used in trawl nets. The
92 estimated 15 to 51 trillion plastic particles weighing between 93 and 236 thousand tons floating in
93 the ocean is based on trawl net data (van Sebille et al., 2015). A recent study showed that trawl
94 net studies could be undersampling particle density by approximately three orders of magnitude
95 (Barrows et al., 2017). This study employed grab sampling, a technique used to sample a limited
96 volume of surface water for microplastic research (Barrows et al., 2017; Miller et al., 2017). Grab
97 samples collect smaller sized particles as well as a greater range of microplastic shapes than a
98 trawl net.

99

100 Understanding the concentrations of microfibers and microplastics is integral to analyzing their
101 potential environmental impact. The last decade of microplastic research has brought attention to
102 the issue and helped decrease knowledge gaps in a new field (Barrows et al., 2017). Under-

reporting of microplastic delays our understanding how the shape and size of microplastics influence their location, important factors for recognizing pollution hot spots and areas of increased biological impacts. Models predicting accumulation at the poles (Isobe et al., 2017; Wilcox et al., 2015) have noted areas of high particle concentrations; this is matched by empirical studies (Bergmann and Klages, 2012; Cózar et al., 2017) but is still lacking for many areas in the Southern Oceans.

Unfortunately, research can be expensive, challenging, time consuming and often seasonally driven, especially at sea. Researchers are increasingly engaging with citizen scientists to help with large scale data collection (Hoellein et al., 2015; McKinley et al., 2016), with numerous projects focusing on marine plastic pollution (Zettler et al., 2017). To date, wide geographic research into plastics and microplastics in the environment has relied heavily on citizen scientist initiatives (Hidalgo-Ruz and Thiel, 2013; Zettler et al., 2017). Using citizen scientists not only allows for wider data collection, but raises awareness outside of the research community and increases engagement with environmental issues (UNEP, 2011; Zettler et al., 2017). Citizen science can also lead to positive changes in policy outcomes, and can be a rigorous process of scientific data collection to help solve global problems (Cigliano et al., 2015; McKinley et al., 2016).

In this study, we use the term ‘microparticle’ to include both microplastics, microfibers and anthropogenic litter of undetermined material type in the size range of 5 mm to 100 μ m. This work started with a focus on microplastics but was expanded to microparticles when it became clear that other types of materials were a significant component of our samples. The term ‘synthetic microfiber’ indicates fibers manufactured from petrochemicals, chemically synthesized or from semi-synthetic cellulosic material (e.g. rayon), and the term ‘non-synthetic microfiber’ refers to fibers made from natural materials and not chemically synthesized, such as cotton or wool. For this study microfibers that appear to be a blend of synthetic and non-synthetic materials are included with the synthetic microfibers.

This study is the most extensive dataset on microparticle contamination in global coastal marine environments. Over five years, we covered a wide geographic distribution and this study is the first to show extensive grab sampling data. The aim is to better understand the global distribution, concentration and type of surface microparticles in the marine environment. This was completed by implementing a citizen science field protocol focused on high quality assurance, sufficient data collection, ease of use and accessibility. Using opportunistic collection

of 1-liter grab samples by citizen scientists, we focused on understudied and often remote ocean regions, including coastlines and the open ocean.

Materials and Methods

Experimental Design

One-liter grab samples were collected from marine surface waters following protocols outlined by Barrows et al. (2017). Sample bottles were triple-rinsed with tap water, sealed, and then triple-rinsed *in situ*. Samples were collected up-current of the citizen scientist and sample bottles were capped underwater immediately following sample collection to reduce air contamination. In the instance the water could not be reached from the sampling platform, a bucket was used to collect surface water; the bottle, cap and bucket were triple-rinsed before sample water was collected. While the citizen scientist stood downwind, sample water was poured into the sample jar until overflowing and capped immediately. Samples were closed tightly, packed and mailed to a laboratory in Stonington, Maine for analysis by trained professionals. 1,628 samples were collected by citizen scientists and processed by three professional scientists from 2013 to 2017. The citizen scientists had a wide range of both scientific expertise (from no previous training to professional scientists), and field experience (basic outdoor competency to professional outdoor athletes). For a citizen scientist to participate in the project, they were required to take an online test to confirm they understood and could follow our sampling procedures.

Citizen scientists collected samples from a diversity of sampling platforms (including wading, and from small and large watercraft) and sampling locations (rocky and sandy shorelines, offshore, estuaries, remote and urban). They were asked to record standard field sampling data about the sampling site and time. Citizen scientists recorded data in a smart phone app, as well as on a hard copy data sheet. As a measure of quality assurance, collectors answered protocol adherence questions regarding their sampling technique after collecting their samples (e.g. did you cap your bottle underwater?). Citizen scientists were also asked to submit photos of the clothing they wore while sampling, which were later used by researchers to determine potential sample contamination.

Laboratory Analysis

Samples were processed by vacuum filtration and particles were counted under a stereo microscope (Barrows et al., 2017; Hidalgo-Ruz et al., 2012). The samples were vacuumed filtered over a gridded 0.45 μm filter (Whatman mixed cellulose nitrate, 47 mm diameter, GE Life

Sciences). Water volume was measured and recorded at time of filtration. Before filtration began, lab surfaces were wiped down with a cellulose sponge to reduce potential contamination. All glassware and tools were triple-rinsed as well as forearms and hands. White 100% cotton lab coats were worn. After filtration, the filters were stored in a triple-rinsed glass petri dish to dry for a minimum of 24 hours.

Filters were counted under a stereo microscope at 45× magnification. Particles were identified based on a lack of cellular structures and, in the case of microfibers and microbeads, a uniform shape (Hidalgo-Ruz et al., 2012). If the piece could not be confirmed as microplastic under a stereo microscope, it was examined under a compound microscope and subjected to the hot needle test (De Witte et al., 2014; Devriese et al., 2015). If, after inspection under the compound microscope, there was doubt the piece was microplastic, it was not counted as microplastic but noted as a potential non-synthetic in the laboratory notes. The initial count of microparticles was thought to be 100% microplastics, which was later changed after further analysis. We recognized that a substantial number of the particles we believed were microplastics were in fact non-synthetic microfibers. This creates a potential underestimate of non-synthetic microfibers, which were not thoroughly classified in the initial analysis. Samples containing non-synthetic fibers were noted during laboratory analysis (n= 56 or 4% of the samples). These fibers were not included in the final results because although material analysis indicated we misidentified a portion of non-synthetics as synthetics, quantity, size class and color were not always recorded. Microparticles were categorized by shape (round, fiber, fragment), color (blue, red, transparent, black, other), and from December 2015 till the end of the study by size (0.1 – 1.5 mm, 1.6 – 3.1 mm, 3.2 – 5 mm, and 5.1 – 9.6 mm). These size classes were chosen based on the filter grid length.

Laboratory Controls

To reduce airborne contamination, the laboratory floor and surfaces were vacuumed a minimum of once a week. Lab water and air blanks were run during sample handling to determine any possible lab contamination. Before filtering, a lab tap water blank was vacuum filtered. A blank was also run of the filtrate used to rinse the sample bottle and filtration apparatus; the volume varied from 0.25 L to over 1.0 L. During filtering, a filter was exposed for 30 seconds to mimic the maximum amount of time the sample could have been exposed when transferred from the sample bottle to the filtration apparatus. When filters were open under the stereo microscope for counting, an air exposure blank was placed next to the microscope.

We conducted 265 water blanks and 126 air blanks over the project duration. There was an average microplastic contamination of 0.005 pieces per 0.010 L of water and 0.154 pieces per 8 minutes of air exposure from both synthetic and non-synthetic air borne contamination. Since contamination was minimal we did not subtract contamination from the field sample results. See Text. S1 for data and procedure.

Data Analysis

To standardize the slight variation in sample volume the total particles per sample were divided by sample volume. Water samples were always completely filtered, although the target 1 L varied substantially (0.9 L – 3.15 L, median = 1.2 L), so results are reported as the number of particles per liter. Between 2013 and 2017, we processed 1,628 samples. Of the 1,628 samples, 14% were excluded from final analysis due to incomplete field data, poor sample quality or were collected at depth with SCUBA (see Table S2.). Results were not significantly different when combining Q1 (high quality) and Q2 (low quality) data (12.1 ± 0.6 particles L⁻¹, mean $\pm$ SE). The Q2 samples alone had a slightly higher average (13.7 ± 2.6 particles L⁻¹, mean $\pm$ SE). All calculations are based on the remaining samples (n = 1,393). Samples were assigned to a major ocean basin and a regional sea, if applicable (Table S3), based on boundaries from the International Hydrographic Organization and NOAA's National Centers for Environmental Information (IHO, 1953; NOAA, 2015). Samples that fell within 12 nm from land were classified as 'coastal' and those outside of 12 nm were classified as 'open ocean' (UNCLOS, 1982).

Statistical Analysis

Microsoft Excel™ was used to conduct Mann Whitney-U tests, figures, tables, averages and histogram calculations on the data. The map was created using ©Carto mapping platform with Jenks data quantification.

μFT-IR Analysis

From the Quality 1 (Q1) samples containing ≥ 1 particle per liter, 113 particles were randomly selected for micro Fourier Transform-Infrared Spectroscopy (μFT-IR) to determine the microparticle material. In each sample, the first particle encountered on the round, gridded filter was removed for analysis. A minimum of ten pieces were taken from ten different samples for each ocean basin (Table S4), and the remaining pieces were awarded based on the ratio of the number of pieces available from each basin to the total number of microparticles available for

μFT-IR analysis (see Table S4). Samples within each geographic category were picked randomly within the Q1 samples. MicroVision Laboratories analyzed each particle using a μFT-IR (a Bruker LUMOS FT-IR operated in reflectance mode) to identify material type. The microparticle material spectra was compared to known standards to determine material type for the particle. The LUMOS has a spectral range from 7000-600 cm⁻¹ and uses a VCSEL laser with a wavelength of 850 nm. The instrument was operated using OPUS software.

Results

Ocean Basins and Regions

Worldwide marine surface waters contain 11.8 ± 0.6 particles L⁻¹ (mean $\pm$ SE) (n = 1,393). Ninety percent of the samples contained microparticles. The Arctic (n = 37) and Southern oceans (n = 14) contained the highest surface water average of 31.3 ± 6.5 and 15.4 ± 8.1 particles L⁻¹ (mean $\pm$ SE), respectively. The Atlantic Ocean contained a higher average (13.4 ± 0.9 particles L⁻¹, mean $\pm$ SE) than the Pacific Ocean (7.0 ± 0.8 particles L⁻¹, mean $\pm$ SE). The Indian Ocean contained the lowest average (4.2 ± 1.2 particles L⁻¹, mean $\pm$ SE), compared to the other ocean basins (Fig. 1).

Across all ocean basins, open ocean samples contained a higher average (17.9 ± 1.1 particles L⁻¹, mean $\pm$ SE) (n = 685) than coastal samples (5.9 ± 0.6 particles L⁻¹, mean $\pm$ SE) (n = 708). This nearly 3-fold increase in particle concentration was highly significant despite a diverse range of particle densities in both sample groups (Mann-Whitney U-test P < 0.001, Fig. 2.). The distribution of particle sizes was similar in coastal and open ocean samples. The smallest particle size class (0.1 - 1.5 mm) was predominate along the coast (79%) and open ocean (74%) (Fig. S2.). The percentage of samples containing particles and average particles per liter decreased with increasing size class in both coastal and open ocean samples.

The pattern of significantly higher densities of particles in the open ocean compared with coastal waters was also seen when individual ocean basins were examined separately (Fig. 3). Across ocean basins, there was an increase in microparticles ranging from a 14% increase in the Southern Ocean to a nearly 5-fold increase in microparticles in open ocean versus coastal water for the Indian Ocean (Fig. 3).

The Atlantic and Pacific Ocean samples were further divided into regions based on oceanographic boundaries (Table 1). Within the Pacific Ocean, open ocean Southeast Asian waters had the

highest average, 19.1 ± 12.2 particles L^{-1} (mean $\pm$ SE) and open ocean sites that did not fall into a specific region were the next highest with 18.4 ± 3.5 particles L^{-1} (mean $\pm$ SE). Within the Atlantic, open ocean sites that did not fall into a specific region had the highest average of 18.0 ± 1.2 particles L^{-1} (mean $\pm$ SE) with coastal Caribbean and Gulf of Maine containing an average of 9.9 ± 3.6 particles L^{-1} and 7.6 ± 2.4 particles L^{-1} , respectively (mean $\pm$ SE) (Table 1). There was a significant difference between coastal and open ocean samples within each ocean (Mann Whitney-U: Pacific $P < 0.001$, Atlantic $P < 0.001$) and when comparing the coastal samples to open ocean samples from the combined oceans ($P = < 0.001$). For additional comparison, samples within the Atlantic and Pacific Oceans were separated north and south of the equator. Northern samples in both the Atlantic and Pacific had a higher particle average than the less intensively sampled southern latitudes (Fig. S1).

Particle Classification

Microfibers were the dominant particle shape (91%) with fragments making up the remaining 9%. Clear/transparent and blue colored particles comprised 47% and 29% of the colors quantified, respectively. The remaining particles were quantified as either black (9%), 'other color' (8%), or red (7%). Particles were predominantly 0.1 - 1.5 mm in length (77%). The other particles were 15% 1.6 - 3.1 mm and 8% 3.2- 5 mm (Fig. S2).

Spectral Analysis

Through μ FT-IR we determined the material type of 113 pieces visually identified as plastic. The pieces were categorized as 96% fibers and 4% fragments. The analysis classified $69\% \pm 4.35\%$ (mean $\pm$ SE) of the samples as synthetic or semi-synthetic, 31% as non-synthetic. Unknown cellulosic material made up 7% of the samples and could not be identified further due to a weak spectral match. Since it they were confirmed as cellulose but displayed different spectra to the semi-synthetic cellulose we grouped them with the non-synthetics. The non-synthetic pieces were classified as cotton, wool or unknown cellulose ($n = 16$, $n = 11$, $n = 8$, respectively). The synthetic pieces were characterized as a wide range of polymers, with polyester predominant (Table 2). All fragments were identified as synthetic. Every ocean basin, coastal and open ocean site had a higher percentage of synthetic particles than non-synthetic, except on the coastal Atlantic (Table S1). Synthetics were characterized in 68% of the coastal samples and 70% of the open ocean samples (Table 2).

Atlantic Rally for Cruisers Sample Subset

The largest subset of samples in this study (more than 2/3 of the open ocean samples) came from a collaborative effort during a trans-Atlantic boat race. In November and December of 2014, 88 boats collected 473 samples while participating in the Atlantic Rally for Cruisers (ARC) race from Las Palmas, Canary Islands to Rodney Bay, St. Lucia. The race spanned more than 2,700 nautical miles, with the majority of the race taking place on the open ocean. The 465 open ocean samples accounted for 69% of the total open ocean samples in this study. This raised the possible concern that if these samples had a bias for higher particle counts, it might be driving the coastal/open ocean difference we had observed. We explored this possibility by dividing the data into two subsets, the ARC data and the remaining samples (Table 3). Both subsamples showed similar patterns, ARC coastal samples contained 3.2 ± 1.2 and 17.6 ± 1.3 particles L^{-1} (mean $\pm$ SE) in open ocean samples. These results are similar to what is found in the Atlantic Ocean ($n = 429$), when removing the ARC subset with 6.1 ± 1.0 and 16.7 ± 2.6 particles L^{-1} in coastal and open ocean samples, respectively. The coastal and open ocean trend is also reflected in the global dataset (without the ARC samples $n = 920$) with 6.0 ± 0.6 particles L^{-1} in coastal samples and 18.5 ± 2.0 particles L^{-1} (mean $\pm$ SE) in the open ocean (Table 3).

Citizen scientists collected 15 samples using SCUBA, between 5 and 18 meters depth in the coastal Atlantic, Pacific and Indian Oceans. The samples contained an average of 16.5 ± 2.8 particles L^{-1} (mean $\pm$ SE) but were not included in the data summary, since all other samples from this study were surface samples.

Discussion

Microparticle Distribution in the Oceans

Our study shows the prevalence of small-sized synthetic and non-synthetic microparticles (11.8 ± 0.6 particles L^{-1} , mean $\pm$ SE) throughout the world's marine surface waters with several robust geographic patterns that have been reported in other studies (Cózar et al., 2017; van Sebille et al., 2015; Wilcox et al., 2015). There is a consistent increase in the density of microparticles in the open ocean surface waters compared to coastal samples. In addition, some ocean basins appear to have higher densities of surface microparticles, with the Arctic and Southern Ocean higher than other ocean basins. This has been theoretically predicted (Eriksen et al., 2014; Wilcox et al., 2015) and empirically documented in other studies (Bergmann et al., 2017; Cózar et al., 2017; Isobe et al., 2017; Waller et al., 2017). Microfibers comprised 91% of the particles quantified in this study. Our most surprising result was the high percentage of fibers that, initially identified as microplastic, were instead classified as semi-synthetic or non-synthetic fibers through more

detailed analysis (Table 2). This indicates that semi-synthetic and non-synthetic microfibers could be a significant and overlooked pollutant in global marine surface waters.

There are higher concentrations of microparticles in open ocean environments than along the coast. Our results show this consistently across all the ocean basins. The large subset of ARC data also reflects this trend. Although the majority (~80%) of plastic pollution is estimated to come from land (Andrady, 2011) and is present in coastal waters, this study, along with others supports the hypothesis that currents are constantly moving these particles into the open ocean and to higher latitudes (Bergmann et al., 2017; C3zar et al., 2017; Eriksen et al., 2013b; Law et al., 2014). Conversely, at 4.5-meter depth in the Northeast Pacific Ocean, Desforge et al. (2014) found up to 27x more particles along the coast than the open ocean, using vertical net tows (236 μ m mesh). Interestingly, the Desforge et al. (2014) study showed particle size increased when using distance from shore as a continuous variable. The conflicting study results show that it may be difficult to compare studies based on tow nets with our grab samples. It remains unclear if these dissimilarities are a result of a methodologically driven difference, or if it is due to regional fluxes of particle introduction and movement, leading to different patterns in the coastal to open ocean gradient.

Our findings are consistent with global microplastic accumulation zone model predictions [e.g. (C3zar et al., 2017; Eriksen et al., 2014; van Sebille et al., 2015)] with the exception of the Mediterranean Sea, where our estimates are based on a very small sample size and may not be representative of the region. The open ocean and poles appear to have sequestered and trapped plastic for over half a century, and demonstrate that not only plastics, but semi-synthetic and non-synthetic microfibers are polluting the environment. Our findings of high contamination in Southeast Asian waters is not unexpected due to the proximity to some of the world's top polluting countries (Jambeck et al., 2015), and the many islands, seas and currents of the region potentially slow the movement of plastic into the open ocean environment (Isobe et al., 2014).

After finding less plastic in surface trawls than predicted by models, Eriksen et al. (2014) and C3zar et al. (2014) suggested that particles < 4.75 mm are somehow misplaced from the ocean's surface. Reisser et al. (2014) and Kooi et al. (2017) have demonstrated that some of the "missing" plastic may be just below the depth sampled by a trawl. The higher average number of particles found in our depth samples further reflect this concept. Smaller sized microplastics (< 1 mm) are often encountered in animal ingestion studies (Cole et al., 2011; De Witte et al., 2014;

Desforges et al., 2015; Li et al., 2016; Li et al., 2015; Lusher et al., 2013; Van Cauwenberghe and Janssen, 2014), indicating that a portion of microplastics may be inside of marine animals. Our data suggest an additional explanation, that much of the smaller-sized microparticles, especially microfibers, are found at the surface (and potentially at depth) but are being incompletely collected by the trawl samples that dominate the literature and have historically provided estimates on microplastic concentration. Even the highest microplastic values captured in the van Sabille et al. (van Sebille et al., 2015) 1 – 5 mm model solution (0.01 particles L⁻¹) are approximately two orders of magnitude less than our average particles between 1.5 – 9.6 mm and three orders of magnitude less when including the smallest size class (0.1 – 1.5 mm). A previous study, with concurrent collecting of trawl net and grab samples, reported a concentration increase of approximately three orders of magnitude with grab samples (Barrows et al., 2017). Grab sampling shows what may be a significant concentration of smaller-sized synthetic, semi-synthetic and non-synthetic microparticles on the sea surface and surface microlayer (Song et al., 2014). This general use of trawl nets may also be why there is underreporting of small semi-synthetic and non-synthetics in the literature, since many small particles and fibers may not be captured by a net. Although, given our initial misidentification of non-synthetic fibers as synthetic, we suspect that a substantial amount of reporting may include non-synthetic fibers misidentified as microplastics.

The small water volume collected using the grab sample technique can lead to high data variability, even within a given place and time, but we show that what it lacks in precision it makes up for in accuracy. Its strength as a sampling technique lies in the ability to control for field and laboratory contamination, its accessibility for opportunistic sampling and its capacity for capturing micro- to nano- sized plastics (Barrows et al., 2017). Although grab samples provide a precise snapshot of microparticle pollution, it is unknown how illustrative it is of a larger region. Side-by-side grab sampling should be considered for future trawl net studies in order to capture a representative sample of diverse particles, inclusive of those smaller than the net mesh size.

Finding a higher concentration of smaller-sized microparticles along the coast and in the open ocean than many studies (Eriksen et al., 2014; Law et al., 2010) is almost certainly a function of the grab sampling method. The method is only restricted by the filter pore size (0.45 µm) in the laboratory rather than the larger size mesh (335 µm) of a trawl net. Some waste water treatment plants (WWTPs) can capture a large amount of microplastic and microfibers (Carr et al., 2016; Mintenig et al., 2017; Talvitie et al., 2017), but still a high proportion appear to slip through,

making them a significant pollutant source (Browne et al., 2011; Eriksen et al., 2013a; Estahbanati and Fahrenfeld, 2016; Hartline, 2016; Mason et al., 2016; Pirc et al., 2016). Particles < 300 μm are less likely to be captured within a WWTP facility (Magnusson and Norén, 2014) or captured by trawl net studies. This may indicate that although severe degradation of larger particles can happen along the coast, many secondary microplastics and non-synthetic fibers may already be quite small (< 1 mm) when entering the marine environment. Their small size and predominately fiber shape suggests they are being underestimated in most studies.

Atmospheric shedding is another potential source of microfiber pollution and could represent a significant percentage of the microfibers captured in this study. Dris et al. (2016) discovered passive atmospheric fallout of between 2 and 355 particles per square meter per day on Parisian rooftops. Fibers were the dominant particle type and 50% were non-synthetic (Dris et al., 2016). Initial results show that airborne transport of both synthetic and non-synthetic fibers may be a significant means of pollution to remote areas of the world. Finding fibers in the open ocean may represent some combination of water and wind transport of land-based or marine-based shed fibers.

Material Characterization

This study initially focused on microplastics, and we attempted to exclude non-synthetic particles through visual microplastic characterization. This method excluded just a few particles, with an estimation of 4% of the samples containing non-synthetics. However, in the chemical verification process, we found 31% of the particles initially identified as microplastic were characterized as non-synthetic fibers, including cotton, wool and cellulose. Even following strict protocols (Barrows et al., 2017; Hidalgo-Ruz et al., 2012; Norén, 2007) when identifying particles as microplastics, over a quarter of our particles were misidentified based on the $\mu\text{FT-IR}$ analysis. If this result is substantiated in future studies, it suggests that other researchers are under-sampling or misidentifying non-synthetic fibers.

We were intrigued to find that some of the plastics characterized in our study are typically denser than seawater. The location of microplastic in the water column appears to be influenced by a complex and frequently changing interaction between polymer composition (density, shape and texture), and variation in the marine environment (fouling organisms, temperature, surface circulation, wind, water density and salinity (Barnes et al., 2009; Cole et al., 2011; Kooi et al., 2017; Thompson et al., 2004). While many low-density plastics occur at the sea surface (e.g. LD

polyethylene, polypropylene), similar types of polymers have been found in the water column and sediments, demonstrating that the initial specific density of the plastic may not be a major influencing factor in where it occurs in the marine environment (Kooi et al., 2017; Thompson et al., 2004). Tidal and wind-induced vertical mixing may suspend high-density polymers (e.g. Polyvinylchloride, polyester) at the sea surface or throughout the water column (Isobe et al., 2014; Kukulka et al., 2012; Law et al., 2014; Reisser et al., 2014). Conversely, low-density polymers may become increasingly dense through biofouling (Kooi et al., 2017; Morét-Ferguson et al., 2010). Biofouled plastics sink to a level in the water column where defouling may occur due to reduced light penetration, grazing, ingestion or water chemistry (Cózar et al., 2014; Ye and Andrady, 1991). Recent modeling shows that microplastic may perpetually oscillate vertically (Kooi et al., 2017) which could mean continuous interaction with aquatic animals.

We chose μ FT-IR analysis after very limited success with Raman spectroscopy characterization of fibers < 1.5 mm in size. The degraded and generally weathered state of the particles increase the uncertainty of accurate material characterization for both spectroscopic analysis techniques. Despite the degraded state of some particles, all were characterized through matching reference spectra peaks. Even though we only had a small sample size, we were surprised at the proportion of particles present in the samples that were identified as non-synthetic, especially those from remote locations. This further reinforces the acknowledged need for additional analysis to be taken during sample quantification, both to help discern between synthetic, semi-synthetic and non-synthetic particles and to confirm the high concentrations of non-synthetic microfibers found in this study. Other low-cost techniques could be considered for distinguishing non-synthetic fibers, such as polarized light microscopy (Santana et al., 2016). Recording the presence of semi-synthetic and non-synthetic particles should also be added to any data collection, as we do not yet know the environmental implications of this emerging pollutant.

There was no substantially higher proportion of synthetic microfibers in any one coastal or open ocean area, or in any particular ocean basin. Therefore, the general trend within our data set appears to apply to all microparticles. Our expectation would have been that non-plastic microfibers would have degraded faster than microplastics, so that for example, in the open ocean we would have expected a higher proportion of microplastics, but the proportions remained relatively stable and we did not see any trends in our relatively small sample size. Interestingly, our results are similar to the current global production of textile fibers at 60% synthetic fibers, 30% cotton and 10% other material (Carr, 2017). The study shows a significant number of

particles make their way to the open ocean and higher latitudes, where degradation rates may be slowed, resulting in long residence time. Non-synthetic and semi-synthetic anthropogenic particles and fibers may pose a new and mostly unconsidered environmental and biological impact.

The percentage of cotton, wool and cellulose discovered across the ocean samples merits further verification and research into both the degradation time in aquatic ecosystems, and environmental health impacts of dyes and other additives used in textile production. The distinct surface properties of non-synthetic fibers cause different chemical sorption behaviors than those of plastic particles (Ladewig et al., 2015; Sillanpää and Sainio, 2017). Many dyes and chemicals used in natural and synthetic textile production have been shown to be carcinogenic to animals (Lithner et al., 2009; Lithner et al., 2011; Remy et al., 2015). Degradation of non-synthetic particles will result in dispersion of these harmful additives into the environment (Remy et al., 2015) and may even occasionally be completely digested when ingested (Zhao et al., 2016). There is a clear lack of research into the field of non-synthetic and semi-synthetic anthropogenic particles in the environment, although they have been found as an airborne contaminate (Dris et al., 2017; Dris et al., 2016), in rivers (Hoellein et al., 2014; McCormick, 2015; McCormick and Hoellein, 2016; McCormick et al., 2016; Miller et al., 2017) , as well as reported to be ingested by birds, fish and macrofauna (GESAMP, 2015; Lusher et al., 2015; Lusher et al., 2013; Remy et al., 2015; Rochman et al., 2015; Wilcox et al., 2015; Zhao et al., 2016). Even though natural and biodegradable materials are predicted to break down relatively quickly due to their chemical structure, preliminary studies (Bagheri et al., 2017; Balestri et al., 2017), including this one, show they are distributed in marine environments. Particle persistence in the environment increases the possibility of biota interaction over a longer period of time and larger geographic area.

Citizen Science, Quality Assurance, and the Collection of Global Data

This study successfully harnessed the power of citizen science to collect high-quality data on a global scale. Coupling the sampling methodology with professional laboratory services led to a large, wide-ranging dataset that provides broader insight into oceanic microparticle distribution than previous studies. Through this research we have gained additional knowledge on microplastic and non-synthetic particle types and distribution in marine surface waters. There are two limits to this field methodology: the volume collected and in the case of this study, the ability to resample at the same site. At the same time, this methodology allows for a robust and consistent dataset that shows global and regional patterns. Continued focus on small-sized

particles is necessary for comprehending ecosystem and population level impacts of leached chemicals associated with synthetic and non-synthetic particles in aquatic environments. Our research highlights the prevalence of anthropogenic microparticles in surface water across the world's oceans.

Conclusions

These results confirm an alarming trend in recent microparticle data: more areas are showing higher particle densities, especially those far from pollution sources, implying a long residency time of both synthetic and non-synthetic materials. Samples came from understudied ocean regions, some of which are emerging as areas of concentrated floating plastic and anthropogenic debris, influenced by distant waste mismanagement and/or airborne particles. Continuing to fill knowledge gaps on microparticle type, shape, and size in remote ocean areas will drive more accurate oceanographic models of pollution accumulation zones. Incorporation of smaller-sized particles in these models, which has previously been lacking, will help us to better understand potential fate and transformation of microparticles in the marine environment.

529 Fig. 1. Average particles L⁻¹ of surface water for each major ocean basin. Number of samples
 530 shown above each bar. Error bars show standard error.
 531

532
 533 Fig. 2. Histogram of binned particle concentrations in coastal (n = 708) and open ocean (n = 685)
 534 samples. Overall median particle concentration was four. Note that the size of bins changes at 20
 535 particles per liter. Above that, density categories were binned in ranges of 20 particles, below that
 536 they were binned in ranges of 2 particles, with a unique bin for zero particles.
 537

Fig. 3. Average particles L⁻¹ in coastal (within 12 nautical miles of land) and open ocean (12 nautical miles outside of shoreline zone) surface samples in the five major oceans. Error bars show standard error. Photo of blue microfiber on filter at 40x magnification.

Table 1. Sample regions within the Atlantic and Pacific Oceans (mean $\pm$ SE). There is a significant difference between coastal and open ocean samples within each ocean (Mann Whitney-U: Pacific $p = 7.24 \times 10^{-8}$, Atlantic $p = 1.69 \times 10^{-32}$) and across both oceans ($p = 2.63 \times 10^{-54}$). Total number of samples taken in each region are shown in parentheses.

Ocean	Region	Coastal particles L ⁻¹	Open ocean particles L ⁻¹	Combined particles L ⁻¹
Pacific	Central America	4.4 $\pm$ 0.7 (49)	3.1 $\pm$ 1.8 (5)	4.3 $\pm$ 0.7 (54)
	Gulf of Alaska	8.2 (52)	-	8.2 $\pm$ 1.8 (52)
	SE Alaska	5.6 (34)	-	5.6 $\pm$ 1.3 (34)
	SE Asia	5.3 $\pm$ 1.4 (33)	19.1 $\pm$ 12.2 (9)	8.3 $\pm$ 2.9 (42)
	Pacific Ocean- <i>unspecified</i>	2.2 $\pm$ 0.4 (147)	18.4 $\pm$ 3.5 (68)	7.3 $\pm$ 1.2 (215)
Atlantic	Caribbean	9.9 $\pm$ 3.6 (83)	6 $\pm$ 3.7 (24)	9.0 $\pm$ 2.9 (107)
	Gulf of Maine	7.6 (6)	-	7.6 $\pm$ 2.4 (6)
	Gulf of Mexico	3.0 (13)	-	3.0 $\pm$ 1.5 (13)
	Mediterranean	2.1 $\pm$ 0.8 (7)	9.0 $\pm$ 1.4 (6)	5.3 $\pm$ 1.2 (13)
	Atlantic Ocean- <i>unspecified</i>	4.7 $\pm$ 0.4 (204)	18.0 $\pm$ 1.2 (559)	14.4 $\pm$ 0.9 (763)

548
549

550
551
552
553
554
555
556

557
558

Table 2. Material type in coastal and open ocean samples as determined by μ FT-IR.

Material Type		Coastal	Open Ocean
Non-synthetic		32%	30%
Synthetic		68%	70%
Non-synthetic	Cotton	5	11
	Wool	10	1
	Unknown Cellulose	5	3
Synthetic	Acrylonitrile	2	3
	Synthetic/Cotton or Cellulose Blend	8	1
	PET	7	4
	Polyester	8	15
	Polyamide/Nylon	3	4
	Polypropylene	3	2
	Polyethylene	2	1
	PVC	1	0
	Semi-Synthetic Cellulose	9	5
Total number of samples analyzed		63	50

Table 3. Comparison of ARC data to the coastal and open ocean Atlantic Ocean and global data. ARC samples contributed a significant portion to the number of open ocean samples collected in the study. Comparative results show that ARC sampling results showed similar particle distribution to the Atlantic Ocean and the global samples.

Dataset	Location	Particle L ⁻¹ (mean $\pm$ SE)	No. of samples	Total No. of samples
Atlantic Rally Cruisers (ARC)	Coastal	3.2 $\pm$ 1.2	8	473
	Open Ocean	17.6 $\pm$ 1.32	465	
Atlantic Ocean exclusive of ARC	Coastal	6.1 $\pm$ 1.0	305	429
	Open ocean	16.7 $\pm$ 2.6	124	
Global samples exclusive of ARC	Coastal	6.0 $\pm$ 0.6	700	920
	Open ocean	18.5 $\pm$ 2.0	220	
Global samples inclusive of ARC	Coastal	5.9 $\pm$ 0.6	708	1393
	Open Ocean	17.9 $\pm$ 1.1	685	

559

560 **Acknowledgments**

561 **General:** Special thanks to Courtney Neumann, Margie Pfeffer and Samantha Corriveau for
562 unwavering laboratory assistance. Adventure Scientists staff provided exceptional support and
563 kept all things volunteer running smoothly throughout the project duration. Thank you to
564 Benjamin Jackson and Katie Christiansen for manuscript editing and commenting. Thank you
565 Chelsea Rochman for reviewing the manuscript. Most of all, Abigail Barrows would like to
566 thank the hundreds of Adventure Scientists volunteers that persevered through difficult field
567 conditions to collect samples for this project. This research would not have been possible without
568 your dedication.

569

570 **Funding:** Maine Space Grant, Patagonia, Norcross Wildlife Foundation, The Dorothy Jordan
571 Chadwick Fund, Charlotte’s Web Foundation, Crutcher Family Foundation, Lindblad
572 Expeditions-National Geographic Fund, Klean Kanteen and Adventure Scientists.

573

574 **Author contributions:**

575 APWB: Project design, sample analysis, data analysis, introduction, results, discussion,
576 supplementary material
577 SEC: Sample processing, manuscript edits, materials and methods
578 CWP: Major editing in all manuscript sections, results

579

580 **Competing interests:** The authors declare no competing interests, financial or otherwise.

581

582

583

584

585

586 **Reference**

587 Andrady, A.L., 2011. Microplastics in the marine environment. Mar Pollut Bull 62, 1596-1605.
588 Arthur, C., Baker, J., Bamford, H., 2009. Proceedings of the international research workshop on
589 the occurrence, effects, and fate of microplastic marine debris, International Research Workshop
590 on the Occurrence, Effects, and Fate of Microplastic Marine Debris. NOAA Technical
591 Memorandum NOS-OR&R-30., Tacoma, Washington, USA.
592 Auta, H.S., Emenike, C.U., Fauziah, S.H., 2017. Distribution and importance of microplastics in
593 the marine environment: A review of the sources, fate, effects, and potential solutions. Environ
594 Int 102, 165-176.

595 Bagheri, A.R., Laforsch, C., Greiner, A., Agarwal, S., 2017. Fate of so-called biodegradable
 596 polymers in seawater and freshwater. *Global Challenges* 1, 1700048.
 597 Balestri, E., Menicagli, V., Vallerini, F., Lardicci, C., 2017. Biodegradable plastic bags on the
 598 seafloor: A future threat for seagrass meadows? *Science of The Total Environment* 605–606, 755-
 599 763.
 600 Barnes, D.K., Galgani, F., Thompson, R.C., Barlaz, M., 2009. Accumulation and fragmentation of
 601 plastic debris in global environments. *Philos Trans R Soc Lond B Biol Sci* 364, 1985-1998.
 602 Barrows, A.P.W., Neumann, C.A., Berger, M.L., Shaw, S.D., 2017. Grab vs. neuston tow net: a
 603 microplastic sampling performance comparison and possible advances in the field. *Analytical*
 604 *Methods* 9, 1446-1453.
 605 Bergmann, M., Klages, M., 2012. Increase of litter at the Arctic deep-sea observatory
 606 HAUSGARTEN. *Marine Pollution Bulletin* 64, 2734-2741.
 607 Bergmann, M., Peeken, I., Beyer, B., Krumpen, T., Primpke, S., Tekman, M.B., Gerds, G., 2017.
 608 Vast quantities of microplastics in Arctic sea ice—a prime temporary sink for plastic litter and a
 609 medium of transport, *Fate and Impact of Microplastics in Marine Ecosystems*. Elsevier, pp. 75-
 610 76.
 611 Boucher, J., Friot, D., 2017. Primary microplastics in the oceans: A global evaluation of sources,
 612 in: João Matos de Sousa, M.P.O., IUCN (Ed.), IUCN, Gland, Switzerland, p. 43.
 613 Browne, M.A., Crump, P., Niven, S.J., Teuten, E., Tonkin, A., Galloway, T., Thompson, R.,
 614 2011. Accumulation of microplastic on shorelines worldwide: sources and sinks. *Environ Sci*
 615 *Technol* 45, 9175-9179.
 616 Carr, S.A., 2017. Sources and dispersive modes of micro-fibers in the environment. *Integrated*
 617 *Environmental Assessment and Management* 13, 466-469.
 618 Carr, S.A., Liu, J., Tesoro, A.G., 2016. Transport and fate of microplastic particles in wastewater
 619 treatment plants. *Water Research* 91, 174-182.
 620 Cigliano, J.A., Meyer, R., Ballard, H.L., Freitag, A., Phillips, T.B., Wasser, A., 2015. Making
 621 marine and coastal citizen science matter. *Ocean & Coastal Management* 115, 77-87.
 622 Cole, M., Lindeque, P., Halsband, C., Galloway, T.S., 2011. Microplastics as contaminants in the
 623 marine environment: a review. *Mar Pollut Bull* 62, 2588-2597.
 624 Cózar, A., Echevarría, F., González-Gordillo, J.I., Irigoien, X., Úbeda, B., Hernández-León, S.,
 625 Palma, Á.T., Navarro, S., García-de-Lomas, J., Ruiz, A., Fernández-de-Puelles, M.L., Duarte,
 626 C.M., 2014. Plastic debris in the open ocean. 111, 10239-10244.
 627 Cózar, A., Martí, E., Duarte, C.M., García-de-Lomas, J., van Sebille, E., Ballatore, T.J., Eguíluz,
 628 V.M., González-Gordillo, J.I., Pedrotti, M.L., Echevarría, F., Troublè, R., Irigoien, X., 2017. The
 629 Arctic Ocean as a dead end for floating plastics in the North Atlantic branch of the Thermohaline
 630 Circulation. *Sci Adv* 3, e1600582.
 631 De Witte, B., Devriese, L., Bekaert, K., Hoffman, S., Vandermeersch, G., Cooreman, K.,
 632 Robbens, J., 2014. Quality assessment of the blue mussel (*Mytilus edulis*): comparison between
 633 commercial and wild types. *Mar Pollut Bull* 85, 146-155.
 634 Desforges, J.P., Galbraith, M., Dangerfield, N., Ross, P.S., 2014. Widespread distribution of
 635 microplastics in subsurface seawater in the NE Pacific Ocean. *Mar Pollut Bull* 79, 94-99.
 636 Desforges, J.P., Galbraith, M., Ross, P.S., 2015. Ingestion of microplastics by zooplankton in the
 637 Northeast Pacific Ocean. *Arch Environ Contam Toxicol* 69, 320-330.
 638 Devriese, L., van der Meulen, M.D., Maes, T., Bekaert, K., Paul-Pont, I., Frère, L., Robbens, J.,
 639 Dick Vethaak, A., 2015. Microplastic contamination in brown shrimp (*Crangon crangon*,
 640 Linnaeus 1758) from coastal waters of the Southern North Sea and Channel area. *Marine*
 641 *Pollution Bulletin* 98, 179-187.
 642 Dris, R., Gasperi, J., Mirande, C., Mandin, C., Guerrouache, M., Langlois, V., Tassin, B., 2017. A
 643 first overview of textile fibers, including microplastics, in indoor and outdoor environments.
 644 *Environ Pollut* 221, 453-458.

645 Dris, R., Gasperi, J., Saad, M., Mirande, C., Tassin, B., 2016. Synthetic fibers in atmospheric
646 fallout: A source of microplastics in the environment? *Mar Pollut Bull* 104, 290-293.

647 Eriksen, M., Lebreton, L.C.M., Carson, H.S., Thiel, M., Moore, C.J., Borerro, J.C., Galgani, F.,
648 Ryan, P.G., Reisser, J., 2014. Plastic Pollution in the World's Oceans: More than 5 Trillion
649 Plastic Pieces Weighing over 250,000 Tons Afloat at Sea *PLoS One* 9, e111913.

650 Eriksen, M., Mason, S., Wilson, S., Box, C., Zellers, A., Edwards, W., Farley, H., Amato, S.,
651 2013a. Microplastic pollution in the surface waters of the Laurentian Great Lakes. *Marine*
652 *Pollution Bulletin* 77, 177-182.

653 Eriksen, M., Maximenko, N., Thiel, M., Cummins, A., Lattin, G., Wilson, S., Hafner, J., Zellers,
654 A., Rifman, S., 2013b. Plastic pollution in the South Pacific subtropical gyre. *Marine Pollution*
655 *Bulletin* 68, 71-76.

656 Estahbanati, S., Fahrenfeld, N.L., 2016. Influence of wastewater treatment plant discharges on
657 microplastic concentrations in surface water. *Chemosphere* 162, 277-284.

658 Galgani, F., Hanke, G., Maes, T., 2015. Global distribution, composition and abundance of
659 marine litter, in: Bergmann, M., Gutow, L., Klages, M. (Eds.), *Marine Anthropogenic Litter*.
660 Springer International Publishing, Cham, pp. 29-56.

661 Gall, S.C., Thompson, R.C., 2015. The impact of debris on marine life. *Marine Pollution Bulletin*
662 92, 170-179.

663 GESAMP, 2015. Sources, fate and effects of microplastic in the marine environment: A global
664 assessment, in: Kershaw, P.J. (Ed.), *GESAMP Reports and Studies*. GESAMP, 4 Albert
665 Embankment, London SE1 7SR, p. 96.

666 Hartline, N.L., 2016. Microfiber masses recovered from conventional machine washing of new or
667 aged garments. *Environmental Science & Technology* 50, 11532-11538.

668 Hidalgo-Ruz, V., Gutow, L., Thompson, R.C., Thiel, M., 2012. Microplastics in the marine
669 environment: a review of the methods used for identification and quantification. *Environmental*
670 *Science & Technology* 46, 3060-3075.

671 Hidalgo-Ruz, V., Thiel, M., 2013. Distribution and abundance of small plastic debris on beaches
672 in the SE Pacific (Chile): a study supported by a citizen science project. *Mar Environ Res* 87-88,
673 12-18.

674 Hoellein, T.J., Rojas, M., Pink, A., Gasior, J., Kelly, J.J., 2014. Anthropogenic litter in urban
675 freshwater ecosystems: Distribution and microbial interactions. *PLoS One* 9, e98485.

676 Hoellein, T.J., Westhoven, M., Lyandres, O., Cross, J., 2015. Abundance and environmental
677 drivers of anthropogenic litter on 5 Lake Michigan beaches: A study facilitated by citizen science
678 data collection. *Journal of Great Lakes Research* 41, 78-86.

679 IHO, 1953. Limits of oceans and seas, 3rd ed. International Hyrdographic Organization.

680 Isobe, A., Kubo, K., Tamura, Y., Kako, S., Nakashima, E., Fujii, N., 2014. Selective transport of
681 microplastics and mesoplastics by drifting in coastal waters. *Mar Pollut Bull* 89, 324-330.

682 Isobe, A., Uchiyama-Matsumoto, K., Uchida, K., Tokai, T., 2017. Microplastics in the Southern
683 Ocean. *Mar Pollut Bull* 114, 623-626.

684 Jambeck, J.R., Geyer, R., Wilcox, C., Siegler, T.R., Perryman, M., Andrady, A., Narayan, R.,
685 Law, K.L., 2015. Plastic waste inputs from land into the ocean. *Science* 347, 768.

686 Kooi, M., Nes, E.H.v., Scheffer, M., Koelmans, A.A., 2017. Ups and downs in the ocean: Effects
687 of biofouling on vertical transport of microplastics. *Environmental Science & Technology* 51,
688 7963-7971.

689 Kukulka, T., Proskurowski, G., Morét-Ferguson, S., Meyer, D.W., Law, K.L., 2012. The effect of
690 wind mixing on the vertical distribution of buoyant plastic debris. *Geophysical Research Letters*
691 39, n/a-n/a.

692 Ladewig, S.M., Bao, S., Chow, A.T., 2015. Natural fibers: A missing link to chemical pollution
693 dispersion in aquatic environments. *Environmental Science & Technology* 49, 12609-12610.

694 Law, K.L., Moret-Ferguson, S., Maximenko, N.A., Proskurowski, G., Peacock, E.E., Hafner, J.,
 695 Reddy, C.M., 2010. Plastic accumulation in the North Atlantic subtropical gyre. *Science* 329,
 696 1185-1188.

697 Law, K.L., Moret-Ferguson, S.E., Goodwin, D.S., Zettler, E.R., Deforce, E., Kukulka, T.,
 698 Proskurowski, G., 2014. Distribution of surface plastic debris in the eastern Pacific Ocean from
 699 an 11-year data set. *Environ Sci Technol* 48, 4732-4738.

700 Lebreton, L.C.M., van der Zwet, J., Damsteeg, J.-W., Slat, B., Andrady, A., Reisser, J., 2017.
 701 River plastic emissions to the world's oceans. *Nat Commun* 8, 15611.

702 Li, J., Qu, X., Su, L., Zhang, W., Yang, D., Kolandhasamy, P., Li, D., Shi, H., 2016.
 703 Microplastics in mussels along the coastal waters of China. *Environmental Pollution* 214, 177-
 704 184.

705 Li, J., Yang, D., Li, L., Jabeen, K., Shi, H., 2015. Microplastics in commercial bivalves from
 706 China. *Environmental Pollution* 207, 190-195.

707 Lithner, D., Damberg, J., Dave, G., Larsson, Å., 2009. Leachates from plastic consumer products
 708 – Screening for toxicity with *Daphnia magna* 74, 1195-1200.

709 Lithner, D., Larsson, A., Dave, G., 2011. Environmental and health hazard ranking and
 710 assessment of plastic polymers based on chemical composition. *Sci Total Environ* 409, 3309-
 711 3324.

712 Lusher, A.L., Hernandez-Milian, G., O'Brien, J., Berrow, S., O'Connor, I., Officer, R., 2015.
 713 Microplastic and macroplastic ingestion by a deep diving, oceanic cetacean: The True's beaked
 714 whale *Mesoplodon mirus*. *Environmental Pollution* 199, 185-191.

715 Lusher, A.L., McHugh, M., Thompson, R.C., 2013. Occurrence of microplastics in the
 716 gastrointestinal tract of pelagic and demersal fish from the English Channel. *Marine Pollution*
 717 *Bulletin* 67, 94-99.

718 Magnusson, K., Norén, F., 2014. Screening of microplastic particles in and down-stream a
 719 wastewater treatment plant, IVL Report C, p. 22.

720 Mason, S.A., Garneau, D., Sutton, R., Chu, Y., Ehmann, K., Barnes, J., Fink, P., Papazissimos,
 721 D., Rogers, D.L., 2016. Microplastic pollution is widely detected in US municipal wastewater
 722 treatment plant effluent. *Environ Pollut* 218, 1045-1054.

723 McCormick, A., 2015. Anthropogenic litter and microplastic in urban streams: abundance, source,
 724 and fate.

725 McCormick, A.R., Hoellein, T.J., 2016. Anthropogenic litter is abundant, diverse, and mobile in
 726 urban rivers: Insights from cross-ecosystem analyses using ecosystem and community ecology
 727 tools. *Limnology and Oceanography* 61, 1718-1734.

728 McCormick, A.R., Hoellein, T.J., London, M.G., Hittie, J., Scott, J.W., Kelly, J.J., 2016.
 729 Microplastic in surface waters of urban rivers: concentration, sources, and associated bacterial
 730 assemblages. *Ecosphere* 7, e01556-n/a.

731 McKinley, D.C., Miller-Rushing, A.J., Ballard, H.L., Bonney, R., Brown, H., Cook-Patton, S.C.,
 732 Evans, D.M., French, R.A., Parrish, J.K., Phillips, T.B., Ryan, S.F., Shanley, L.A., Shirk, J.L.,
 733 Stepenuck, K.F., Weltzin, J.F., Wiggins, A., Boyle, O.D., Briggs, R.D., Chapin, I.I.S.F., Hewitt,
 734 D.A., Preuss, P.W., Soukup, M.A., 2016. Citizen science can improve conservation science,
 735 natural resource management, and environmental protection. *Biological Conservation* 208, 15-28.

736 Miller, R.Z., Watts, A.J.R., Winslow, B.O., Galloway, T.S., Barrows, A.P.W., 2017. Mountains to
 737 the sea: River study of plastic and non-plastic microfiber pollution in the northeast USA. *Mar*
 738 *Pollut Bull*.

739 Mintenig, S.M., Int-Veen, I., Löder, M.G.J., Primpke, S., Gerdt, G., 2017. Identification of
 740 microplastic in effluents of waste water treatment plants using focal plane array-based micro-
 741 Fourier-transform infrared imaging. *Water Research* 108, 365-372.

742 Morét-Ferguson, S., Law, K.L., Proskurowski, G., Murphy, E.K., Peacock, E.E., Reddy, C.M.,
 743 2010. The size, mass, and composition of plastic debris in the western North Atlantic Ocean.
 744 Marine Pollution Bulletin 60, 1873-1878.
 745 NOAA, 2015. Ocean boundary definitions. US Department of Commerce, NOAA National
 746 Centers for Environmental Information.
 747 Norén, F., 2007. Small plastic particles in coastal Swedish waters KIMO Sweden. KIMO
 748 Sweden Göteborg, Sweden.
 749 O'Neill, C., Hawkes, F.R., Hawkes, D.L., Lourenço, N.D., Pinheiro, H.M., Delée, W., 1999.
 750 Colour in textile effluents - sources, measurement, discharge consents and simulation: a review.
 751 Journal of Chemical Technology & Biotechnology 74, 1009-1018.
 752 Pirc, U., Vidmar, M., Mozer, A., Krzan, A., 2016. Emissions of microplastic fibers from
 753 microfiber fleece during domestic washing. Environmental Science & Pollution Research.
 754 PlasticsEurope, 2016. Plastics—the facts 2016: an analysis of European plastics
 755 production, demand and waste data., in: Europe, P. (Ed.). PlasticsEurope.
 756 Reisser, J., Slat, B., Noble, K., du Plessis, K., Epp, M., Proietti, M., de Sonnevile, J., Becker, T.,
 757 Pattiaratchi, C., 2014. The vertical distribution of buoyant plastics at sea, Biogeosciences
 758 Discussions, pp. 1249-1256.
 759 Remy, F.o., Collard, F., Gilbert, B., Compère, P., Eppe, G., Lepoint, G., 2015. When microplastic
 760 is not plastic: The ingestion of artificial cellulose fibers by macrofauna living in seagrass
 761 macrophytodebris. Environmental Science & Technology.
 762 Rochman, C.M., Tahir, A., Williams, S.L., Baxa, D.V., Lam, R., Miller, J.T., Teh, F.C.,
 763 Werorilangi, S., Teh, S.J., 2015. Anthropogenic debris in seafood: Plastic debris and fibers from
 764 textiles in fish and bivalves sold for human consumption. Sci. Rep. 5, 14340.
 765 Santana, M.F.M., Ascer, L.G., Custódio, M.R., Moreira, F.T., Turra, A., 2016. Microplastic
 766 contamination in natural mussel beds from a Brazilian urbanized coastal region: Rapid evaluation
 767 through bioassessment. Marine Pollution Bulletin 106, 183-189.
 768 Sillanpää, M., Sainio, P., 2017. Release of polyester and cotton fibers from textiles in machine
 769 washings. Environmental Science and Pollution Research 24, 19313-19321.
 770 Song, Y.-K., Han, G.M., Jang, M., 2014. Large accumulation of micro-sized synthetic polymer
 771 particles in the sea surface microlayer. Environ Sci Technol 48, 9014-9021.
 772 Talvitie, J., Mikola, A., Setälä, O., Heinonen, M., Koistinen, A., 2017. How well is microlitter
 773 purified from wastewater? – A detailed study on the stepwise removal of microlitter in a tertiary
 774 level wastewater treatment plant. Water Research 109, 164-172.
 775 Thompson, R.C., Olsen, Y., Mitchell, R.P., Davis, A., Rowland, S.J., John, A.W.G., McGonigle,
 776 D., Russell, A.E., 2004. Lost at sea: Where is all the plastic? Science 304, 838.
 777 UNCLOS, 1982. United Nations Convention on the Law of the Sea, in: Nations, U. (Ed.). United
 778 Nations Treaty Series.
 779 UNEP, 2011. UNEP year book 2011 : emerging issues in our global environment. United Nations
 780 Environment Programme,
 781 Division of Early Warning
 782 Assessment, Nairobi, Kenya.
 783 Van Cauwenberghe, L., Janssen, C.R., 2014. Microplastics in bivalves cultured for human
 784 consumption. Environmental Pollution 193, 65-70.
 785 van Sebille, E., Wilcox, C., Lebreton, L., Maximenko, N., Hardesty, B.D., van Franeker, J.A.,
 786 Eriksen, M., Siegel, D., Galgani, F., Law, K.L., 2015. A global inventory of small floating plastic
 787 debris. Environmental Research Letters 10, 124006.
 788 Verschoor, A.d.P., Leon; Roex, Erwin; Bellert, Bert, 2014. Quick scan and prioritization of
 789 microplastic sources and emissions, in: Environment, M.o.I.a.t. (Ed.). National Institute for Public
 790 Health and the Environment, p. 41.

791 Waller, C.L., Griffiths, H.J., Waluda, C.M., Thorpe, S.E., Loaiza, I., Moreno, B., Pacherres, C.O.,
792 Hughes, K.A., 2017. Microplastics in the Antarctic marine system: An emerging area of research.
793 Science of The Total Environment 598, 220-227.

794 Wilcox, C., Van Seville, E., Hardesty, B.D., 2015. Threat of plastic pollution to seabirds is global,
795 pervasive, and increasing. Proceedings of the National Academy of Sciences Early Edition.

796 Ye, S., Andrady, A.L., 1991. Fouling of floating plastic debris under Biscayne Bay exposure
797 conditions. Marine Pollution Bulletin 22, 608-613.

798 Zettler, E.R., Takada, H., Monteleone, B., Mallos, N., Eriksen, M., Amaral-Zettler, L.A., 2017.
799 Incorporating citizen science to study plastics in the environment. Analytical Methods 9, 1392-
800 1403.

801 Zhao, S., Zhu, L., Li, D., 2016. Microscopic anthropogenic litter in terrestrial birds from
802 Shanghai, China: Not only plastics but also natural fibers. Science of The Total Environment 550,
803 1110-1115.

Supplementary Materials

Text S1. Laboratory QA/QC blanks procedure and calculation.

Fig. S1. Average particles per liter in Atlantic and Pacific Oceans.

Fig. S2. Average particles per size class in coastal and open ocean environments.

Table S1. μ FT-IR analysis by ocean and sample location.

Table S2. Criteria used to evaluate samples on data and sample quality.

Table S3. Major ocean basins and regional seas.

Table S4. Particles selected for μ FT-IR analysis from each ocean basin.

Supplementary Material

Supplementary materials and methods

Text S1. Laboratory QA/QC Blanks Procedure and calculation.

QA/QC- Air Blank

The air blank consists of a Whatman 47mm diameter, 0.45µm gridded filter placed on a triple rinsed glass petri dish. During sample processing and counting two types of air blanks were run on the work bench. One was exposed to the air the same amount of time as the sample (intermittently) and the other was exposed for the duration of processing or counting. 126 air blanks were taken over the duration of the study: 97 intermittent and 29 continuous.

The calculation:

$$313/16208.8 = 0.01931 * 8 = 0.154$$

313 = number of synthetic (37) and non-synthetic (276) particles

16208.8 = total minutes exposed

0.0193 = plastic contamination per minute

8 = average sample exposure time in minutes (7 minutes counting, 30 seconds filtering)

0.154 = Potential piece contamination per sample

QA/QC-Water Blank

During a filtration session, we would process a tap water blank and/or a filtrate blank in the lab to determine potential contamination from water in contact with the sample. The filtrate blank came from sample water that had passed through the 0.45µm filter and collected in a triple rinsed beaker. The filtrate water was placed in a Fisher 1 L squeeze bottle for rinsing down sample jar and flask during filtration. We conducted 265 water blanks of varying volumes of the project duration.

Our calculations, based on the amount of laboratory water in contact with the sample (0.010 L):

$$144/267.075 = 0.539 * 0.010 = 0.005$$

144 = number of plastic pieces

267.075 = volume (L) of water processed

0.0017 = plastic contamination per L

0.010 = maximum volume (L) of laboratory water introduced to the sample

0.005 = potential plastic pieces per 0.010 L

Combining air blank total contamination and water blank total contamination:

$0.154 + 0.005 = \mathbf{0.159}$ pieces of potential plastic contamination per sample

Fig. S1. Average particles $L^{-1} \pm SE$ in Atlantic and Pacific Oceans. Each ocean is separated into coastal and open ocean sites from northern and southern latitudes. Data labels indicated number of samples collected.

Fig. S2. Average particles per size class in coastal and open ocean environments. Note that specific size class was only recorded for 398 samples (coastal n = 327, open ocean n= 71).

Table S1. μ FT-IR analysis by ocean and sample location. Count of non-synthetic and synthetic particles.

Ocean	Location	Non-Synthetic	Synthetic
Arctic	Coastal	2	4
	Open ocean	0	6
Atlantic	Coastal	9	8
	Open ocean	13	22
Indian	Coastal	1	10
	Open ocean	0	2
Pacific	Coastal	4	15
	Open ocean	2	4
Southern	Coastal	4	6
	Open ocean	0	1
Total number of samples		35	78

Sample quality	Data quality
1. Volume more than 0.9 L but no more than 3.15 L	1. All sample location data provided
2. Sample taken in only one bottle - not split between multiple	2. No assumptions had to be made about sample location
3. Low sediment load	3. High quality volunteer self-assessment (4 or above) [if response provided]
4. Sample didn't contain large quantity of aluminum foil*	

Samples taken at > 1 meter depth were also excluded.

Major ocean basin	Regional seas
Atlantic	Caribbean
	Gulf of Mexico
	Mediterranean
Arctic	-
Indian	-
Pacific	Southeast Asia
	Gulf of Alaska
	Southeast Alaska, British Columbia, and WA and OR states
	Western coast of Central and Latin America
Southern	-

Table S4. Number of particles selected for μ FT-IR analysis from each ocean basin.

Ocean Basin	Regional Sea	Number of Grab samples analyzed	Particles selected for μ FT-IR analysis
Arctic	NA	46	12
	NA	873	44
Atlantic	Mediterranean, Black Sea	14	1
	Caribbean	109	6
	Gulf of Mexico	17	1
Indian	NA	54	13
Pacific	NA	259	9
	Gulf of Alaska	60	4
	Coastal waters of Southeast Alaska and British Columbia	40	3
	Southeast Asia	55	7
	Western coast of Central America	64	2
	NA	17	11
	NA	20	0
Total		1628	113

953
954
955
956
957
958
959
960
961
962
963
964
965
966
967
968
969
970
971
972
973
974
975
976