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

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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 ± 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 μFT-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.

1. 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

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(Arthur et al., 2009; Auta et al., 2017; Browne et al., 2011; Dris et al., 2017; Galgani et al., 2015; Verschoor et al., 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; Cole et al., 2011). The relative significance of the two microplastic types are not well studied (Boucher and Friot, 2017), but the majority of particles in surface water appear to be microfibers, a threadlike particle with a length between 100 μm and 5 mm and a width of approximately 1.5 orders of magnitude shorter (Barrows et al., 2017), suggesting secondary microplastics are predominant (Browne et al., 2011; Carr, 2017; Mason et al., 2016).

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

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

Understanding the concentrations of microfibers and microplastics is integral to analyzing their potential environmental impact. The last decade of microplastic research has brought attention to 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−100 μm. 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.

2. Materials and methods

2.1. 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. 1628 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.

2.2. 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 h.

Filters were counted under a stereo microscope at 45x 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.

2.3. 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 s 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 min 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.

2.4. 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 1628 samples. Of the 1628 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 ± SE). The Q2 samples alone had a slightly higher average (13.7 ± 2.6 particles L^{-1}, mean ± SE). All calculations are based on the remaining samples ($n = 1393$). 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).

2.5. 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.

2.6. 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 to 600 cm-1 and uses a VCSEL laser with a wavelength of 850 nm. The instrument was operated using OPUS software.

3. Results

3.1. Ocean basins and regions

Worldwide marine surface waters contain 11.8 ± 0.6 particles L^{-1} (mean ± SE) ($n = 1393$). 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 ± SE), respectively. The Atlantic Ocean contained a higher average (13.4 ± 0.9 particles L^{-1}, mean ± SE) than the Pacific Ocean (7.0 ± 0.8 particles L^{-1}, mean ± SE). The Indian Ocean contained the lowest average (4.2 ± 1.2 particles L^{-1}, mean ± SE), compared to the other ocean basins (Fig. 1).

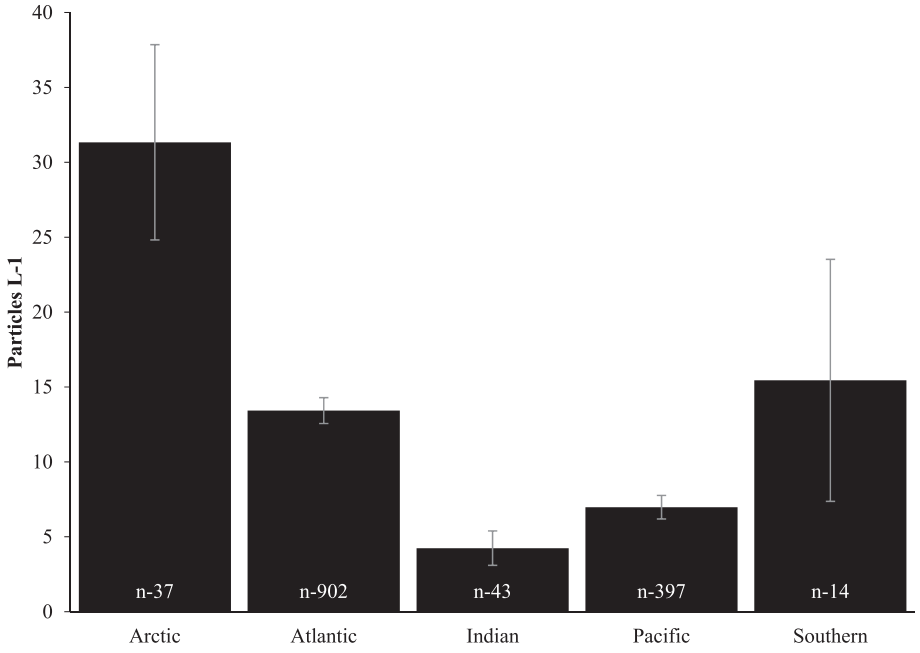

Fig. 1. Average particles L^{-1} of surface water for each major ocean basin. Number of samples shown at bar base. Error bars show standard error.

Across all ocean basins, open ocean samples contained a higher average (17.9 ± 1.1 particles L^{-1}, mean ± SE) (n = 685) than coastal samples (5.9 ± 0.6 particles L^{-1}, mean ± 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.

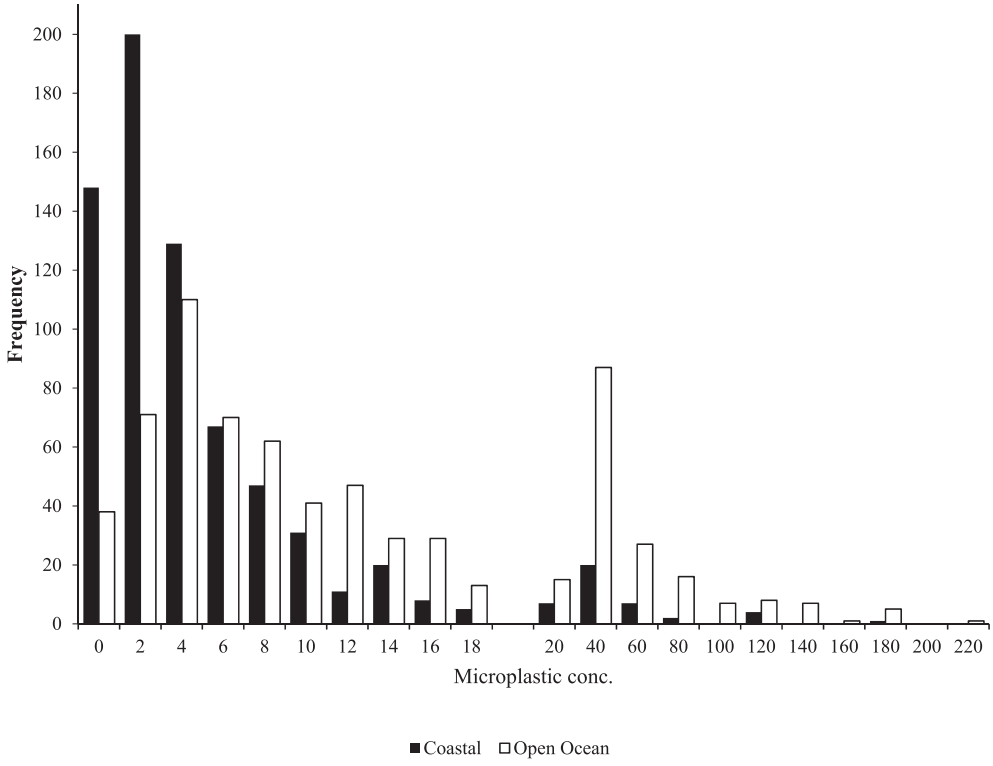

Fig. 2. Histogram of binned particle concentrations in coastal (n = 708) and open ocean (n = 685) samples. Overall median particle concentration was four. Note that the size of bins changes at 20 particles per liter. Above that, density categories were binned in ranges of 20 particles, below that they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

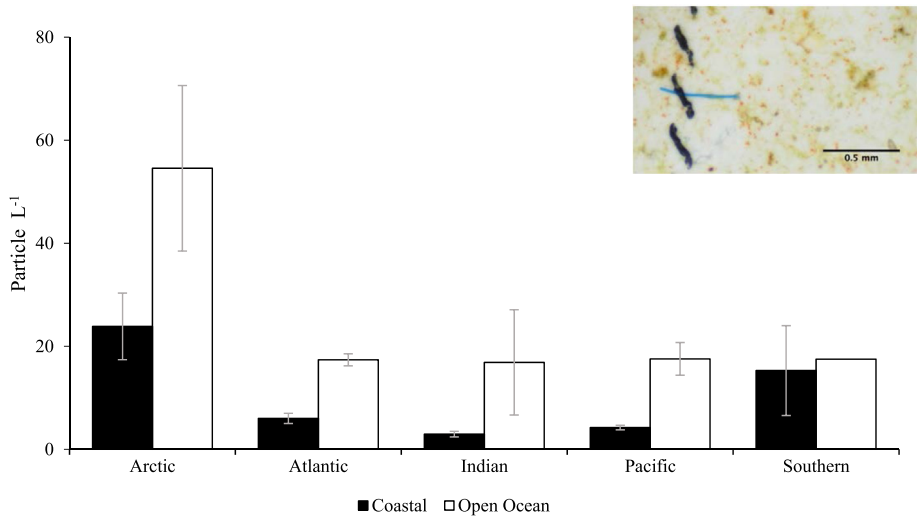

Fig. 3. Average particles L^{-1} in coastal (within 12 nautical miles of land) and open ocean (12 nautical miles outside of shoreline zone) surface samples in the five major oceans. Error bars show standard error. Photo of blue microfiber on filter at 40× magnification. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

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 ± 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 ± 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 ± 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 ± 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).

3.2. 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).

3.3. Spectral analysis

Through μFT-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% ± 4.35% (mean ± 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

Table 1
Sample regions within the Atlantic and Pacific Oceans (mean ± 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 ± 0.7 *(49)*	3.1 ± 1.8 *(5)*	4.3 ± 0.7 *(54)*
	Gulf of Alaska	8.2 *(52)*	–	8.2 ± 1.8 *(52)*
	SE Alaska	5.6 *(34)*	–	5.6 ± 1.3 *(34)*
	SE Asia	5.3 ± *1.4 (33)*	19.1 ± 12.2 *(9)*	8.3 ± 2.9 *(42)*
	Pacific Ocean-*unspecified*	2.2 ± *0.4 (147)*	18.4 ± 3.5 *(68)*	7.3 ± 1.2 *(215)*
Atlantic	Caribbean	9.9 ± 3.6 *(83)*	6 ± 3.7 *(24)*	9.0 ± 2.9 *(107)*
	Gulf of Maine	7.6 *(6)*	–	7.6 ± 2.4 *(6)*
	Gulf of Mexico	3.0 *(13)*	–	3.0 ± 1.5 *(13)*
	Mediterranean	2.1 ± *0.8 (7)*	9.0 ± *1.4 (6)*	5.3 ± 1.2 *(13)*
	Atlantic Ocean-*unspecified*	4.7 ± *0.4 (204)*	18.0 ± *1.2 (559)*	14.4 ± 0.9 *(763)*

Table 2
Material type in coastal and open ocean samples as determined by μFT-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**

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).

3.4. 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 that 2700 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 ± 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 ± SE) in the open ocean (Table 3).

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

4. Discussion

4.1. 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 ± 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

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

hypothesis that currents are constantly moving these particles into the open ocean and to higher latitudes (Bergmann et al., 2017; Cózar et al., 2017; Eriksen et al., 2013b; Law et al., 2014). Conversely, at 4.5-m depth in the Northeast Pacific Ocean, Desforges et al. (2014) found up to 27x more particles along the coast than the open ocean, using vertical net tows (236 μm mesh). Interestingly, the Desforges 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. (Cózar 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 Cózar 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., 2015, 2016; 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 and 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.

4.2. 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 μFT-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 μFT-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, 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., 2016, 2017), 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., 2013, 2015; 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.

4.3. 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.

5. 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.

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Author contributions

APWB: Project design, sample analysis, data analysis, introduction, results, discussion, supplementary material.

SEC: Sample processing, manuscript edits, materials and methods.

CWP: Major editing in all manuscript sections, results.

Conflicts of interest

The authors declare no competing interests, financial or otherwise.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.envpol.2018.02.062.

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