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Abundance and characteristics of microplastics in beach sediments: Insights into microplastic accumulation in northern Gulf of Mexico estuaries

Caitlin C. Wessel [a,b,*], Grant R. Lockridge [a], David Battiste [c], Just Cebrian [a,b]

[a] Dauphin Island Sea Lab, 101 Bienville Blvd., Dauphin Island, AL 36528, USA
[b] University of South Alabama, Marine Science Department, 307 North University Blvd., Mobile, AL 36688, USA
[c] University of South Alabama, Chemistry Department, CHEM0131, 6040 USA Dr., Mobile, AL 36688, USA

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ABSTRACT

Microplastics (plastic debris smaller than 5 mm) represent a growing concern worldwide due to increasing amounts of discarded trash. We investigated microplastic debris on sandy shorelines at seven locations in a northern Gulf of Mexico estuary (Mobile Bay, AL) during the summer of 2014. Microplastics were ubiquitous throughout the area studied at concentrations 66-253× larger than reported for the open ocean. The polymers polypropylene and polyethylene were most abundant, with polystyrene, polyester and aliphatic polyamide also present but in lower quantities. There was a gradient in microplastic abundance, with locations more directly exposed to marine currents and tides having higher microplastic abundance and diversity, as well as a higher contribution by denser polymers (e.g. polyester). These results indicate that microplastic accumulation on shorelines in the northern Gulf of Mexico may be a serious concern, and suggest that exposure to inputs from the Gulf is an important determinant of microplastic abundance.

1. Introduction

Marine debris constitutes, "*any persistent solid material that is manufactured or processed and directly or indirectly, intentionally or unintentionally, disposed of or abandoned into the marine environment or the Great Lakes*," (UNEP, 2009). Anthropogenic litter is found throughout the ocean, even in remote areas far from human contact and obvious sources of pollution (Barnes et al., 2009; Derraik, 2002). The increase in discarded trash, along with very slow degradation rates, is leading to the gradual increase of marine litter found at sea, on the ocean floor, and along the shore. In 2010 between 4.8 and 12.7 million metric tons of plastic litter reached the oceans and an estimated 5 trillion pieces of plastic are currently floating in the ocean (Cozar et al., 2014; Eriksen et al., 2014, Jambeck et al., 2015). Plastics are a diverse group of manufactured materials derived from petrochemicals, and they are lightweight, inexpensive, durable, strong, corrosion resistant, and designed to be disposable. The first plastic polymer (Bakelite) was developed in 1907 and in the 1940s, with the commercialization of plastic products, mass production increased dramatically.

One increasingly abundant type of plastic marine debris is microplastics. They come in a wide range of sizes smaller than 5 mm and have many different shapes (e.g. pellets, fragments, scrubbers; Frias et al., 2010). Microplastic debris has varying levels of buoyancy. Of the 14 different types of plastic compounds that have been found in marine environments, four can have densities lower than freshwater (expanded polystyrene, polyethylene, polypropylene, and polyester) and one has density lower than saltwater (polyamide) (Hidalgo-ruz et al., 2012; Andrady, 2011; Driedger et al., 2015). Plastic debris found in the marine environment can originate from both land and sea, with an estimated 75–90% coming from land-based activities including dumpsites, littering, tourism, fishing, and poor waste management, and 10–25% from sea-based sources, including fishing gear, shipping activities, and dumping (GESAMP, 2010; UNEP, 2005; Sheavly and Register, 2007; Andrady, 2011; Ribic et al., 2011; Mehlhart and Blepp, 2012).

Microplastics are a major environmental problem worldwide. In marine environments microplastics can be transported over long distances by ocean currents and eventually be deposited in coastal habitats such as marshes, seagrass beds, and reefs (Barnes et al., 2009). As petroleum-derived products microplastics can absorb a wide range of hydrophobic toxins including persistent organic pollutants and pharmaceuticals, thereby becoming a transportation vector for these caustic substances (Teuten et al., 2007; Browne et al., 2008, Teuten et al., 2009; Colabuono et al., 2010; Frias et al., 2010; Mato et al., 2001; Bakir et al., 2012; Browne et al., 2013, Bakir et al., 2014). Once transported into coastal habitats, microplastics interact with benthic and pelagic

* Corresponding author.
E-mail address: cwessel@disl.org (C.C. Wessel).

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biota, especially in shallow and well-mixed systems. Upon ingestion microplastic-borne toxins have been shown to desorb from the plastic and accumulate in various tissues and organs causing damaging effects to the organism (Browne et al., 2013; Qiu et al., 2016; Sussarellu et al., 2016).

Studies on microplastics in marine coastal systems have been carried out on both the east and west coasts of North America, the Caribbean, southern Africa, the Mediterranean, Europe, Antarctica and Asia. However, no studies have been carried out in the Gulf of Mexico. Here, we examine the abundance, distribution and composition of microplastics in Mobile Bay, a large estuarine system in the northern Gulf of Mexico. Besides contributing novel information on microplastic occurrence and composition for the Gulf of Mexico, our results provide insights on possible mechanisms that regulate the distribution and accumulation of microplastics along estuarine coastlines.

2. Methods

2.1. Study area

We sampled intertidal sandy sediments in Mobile Bay (Alabama, USA), which is located in the northern Gulf of Mexico (nGoM) and represents the fourth largest estuary in the United States, during the summer season (June–September) at low tide. Salinities within the estuary range from 0 psu (density = 1.00 g/mL) at the northern delta, where five rivers discharge freshwater, to 32 psu (density = 1.03 g/mL) at the mouth (Dauphin Island Sea Lab, 2016). The sampling locations were spread out around the estuary with four in areas primarily influenced by heavy freshwater discharge and forcing (average salinity <0.15 psu), and three in areas dominated by marine tides (average salinity >15 psu, Fig. 1).

2.2. Sample collection

Sampling was carried out along three shoreline stretches at each of the seven locations. Within each stretch, four 0.25 m × 0.25 m quadrats were randomly located along the wrack line (i.e. the line along the shoreline that represents the furthest extent of the most recent high water level) for a total of 12 samples per location. GPS coordinates were obtained for each sampled quadrat and large pieces of natural debris (i.e. seaweed, leaves, wood) was brushed off and removed. Subsequently the top layer of sediment (approximately 3–6 cm) was removed and sieved through a 5 mm sieve into a collection container, items (including plastic) larger than 5 mm were discarded. The collected material (~14 L from each section) was then transported to the lab for processing.

2.3. Microplastic separation

Separation of microplastics from other collected matter was accomplished using a combination of sieving (items between 0.5 and 5 mm), density separation (items between 200 and 500 μm), and visual sorting. Our technique includes novel features that facilitate microplastic isolation. Using a combination of hand sieving and mechanical Ro-tap, all samples were passed through a series of mesh sizes (5 mm, 4 mm, 2 mm, 1 mm, 0.5 mm) and the retained material inspected visually for microplastics. The material that passed through all sieves (<0.5 mm) was retained for density separation.

To carry out the separation of microplastic particles <0.5 mm, we designed a separation process that uses density differences to mechanically separate sand and plastic particles (Fig. 2). The separator was constructed with a series of PVC pipes and connectors. A disk, made from flat stock PVC was secured in between the standard schedule 40 PVC and the bottom tee, served as a barrier between the water reservoir and the sample material. The disk had twenty five, 1-cm holes drilled randomly throughout and 1 mm and 50 μm mesh layers glued to it. The threaded male adaptor on the bottom tee was attached to an ECODIVER 1000 submersible pump that supplied recirculating >35 psu filtered water.

Approximately 3 L of sample material was added at a time through the top tee onto the disk, and the >35 psu water was pumped into the separator through the bottom tee. The flow was adjusted with the ball valve until fluidized sandy sediment was approximately 30 cm from the top of the density separator. Microplastic particles, less dense than sand and the >35 psu water, were carried by the flowing water to the top of the separator. Both microplastic particles and the >35 water exited the separator through the top tee and, after passing through a 200 μm capture sieve, the water was recirculated into the separator. Aeration was provided with a Sweetwater Linear II model SL24 aerator, applied with a 1/4 in clear flexible tubing and airstone that was placed through the top of the separator to approximately 10 cm above the disk. Pneumatic flow was controlled to minimize the amount of sediment material exiting the separator, while still facilitating the upwelling of less dense particles. Each sample was processed for 26 min (the minimum time necessary to capture the most microplastics possible), at

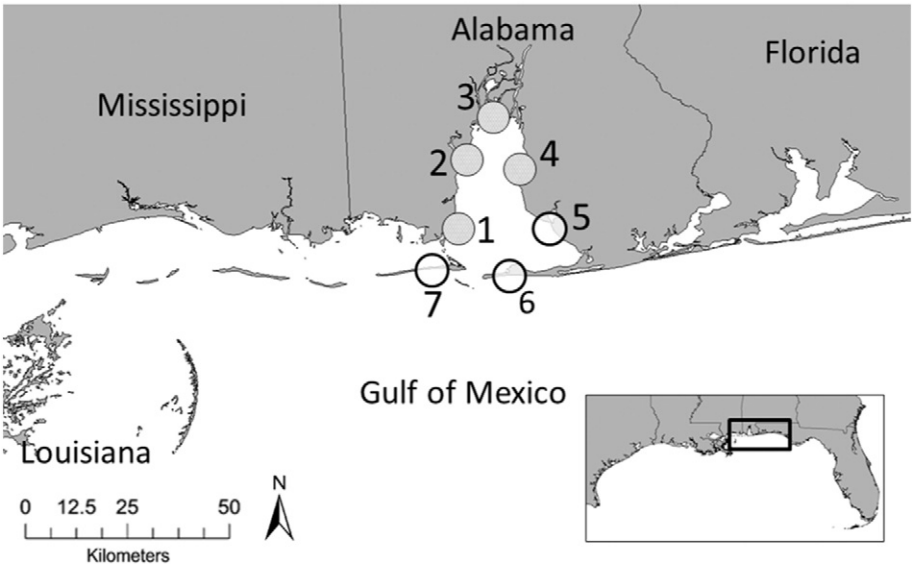

Fig. 1. Study locations around Mobile Bay, AL. Gray dots represent the four areas primarily influenced by riverine outflow and the white dots represent the three in areas heavily influenced by marine tides.

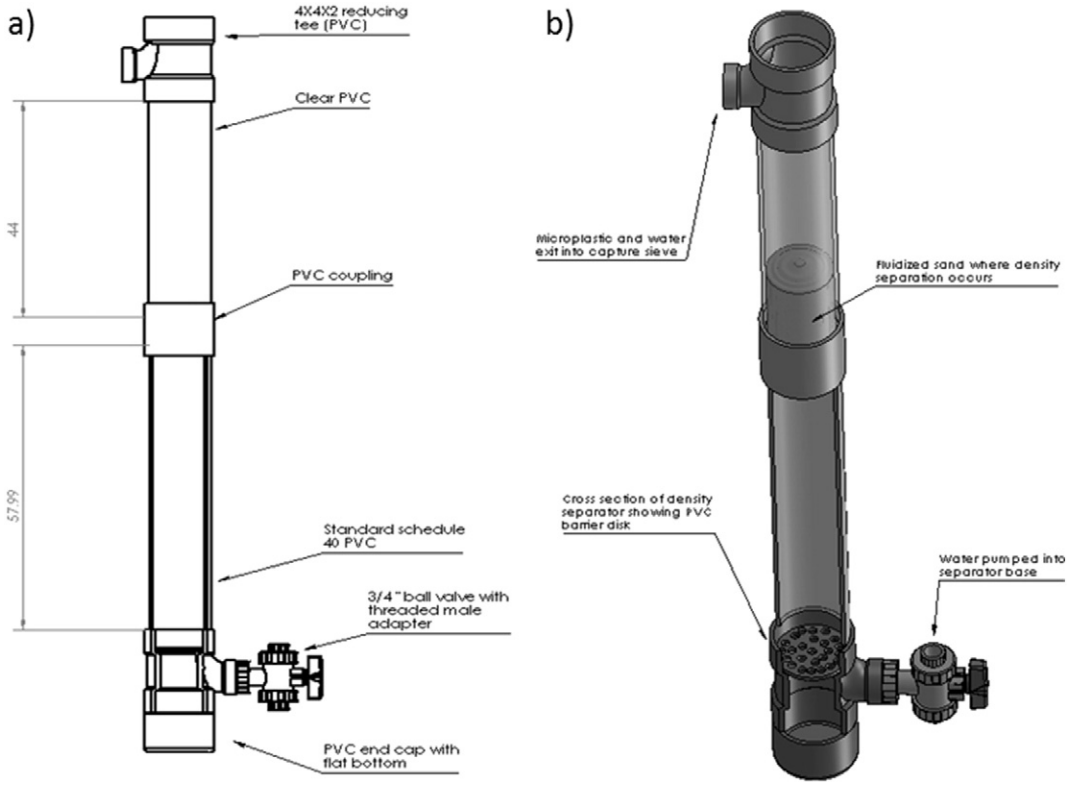

Fig. 2. Representation of the density separator design and setup. a) Part identification and assembly and b) functional depiction identifying internal components and separation process location.

which time water flow was stopped and any particles visible at the exit point of the separator washed into the capture sieve. The sample in the capture sieve was visually examined and sorted to remove non-microplastic materials (i.e. organics, glass). Collected microplastics were dried and stored in a dark climate controlled environment to prevent any further degradation. Densities of microplastics were reported both as items per m^2 and mass per m^2. Efficiency of the density separator was evaluated by adding known quantities of microplastics to sediment samples previously filtered for microplastics. These analyses showed that our separator achieves a mean recovery rate of 97.25% (SD = 2.5).

2.4. Microplastic composition analysis

All of the microplastics from each sample were sorted by size, shape and color, and the length and mass (biofilms, though not obviously present, were also not actively removed and may affect the mass) for each particle recorded before being subjected to attenuated total reflection Fourier transformed infrared spectroscopy (ATR-FTIR) to determine microplastic chemical composition. Plastics are synthetic polymers made from a variety of chemical compounds with different characteristics. Infrared spectroscopy is considered the most reliable method to identify the chemical composition of microplastics (Browne et al., 2010; Hidalgo-ruz et al., 2012). The ATR-FTIR spectrum is compared to a reference database to determine the type of microplastic (e.g. polypropylene, polystyrene, polyethylene). The ATR-FTIR is a single beam, percent transmission technique that runs 16 scans per sample at a resolution of 4.0 cm^{-1} and wavelength range from 4000 to 600 cm^{-1}.

2.5. Statistical analysis

Differences in microplastics abundance and mass were analyzed with nested analysis of variance (ANOVA). The locations examined were divided into two groups based on their marine influence, with stations 1 to 4 primarily influenced by freshwater discharge and forcing, and stations 5 to 7 more largely dominated by marine tides. These locations were nested within the groups, with the shoreline stretches sampled within each location representing the analytical replicate (i.e. the four quadrats sampled within each stretch were averaged into one single replicate). ANOVA assumptions of normality and homogeneity of variance were assessed with the Kolmogorov–Smirnov and Cochran's C-tests, respectively. When necessary, an appropriate transformation was performed before further analysis. When assumptions were not met, the level of significance was set at 0.01 to reduce the possibility of committing Type I error (Underwood, 1997). When assumptions were met a 0.05 significance level was used (Table 1).

3. Results & discussion

In total 84 quadrats were sampled across seven different shorelines located in Mobile Bay, AL. Microplastics were prevalent throughout the bay and were found in all quadrats. This is consistent with results from other regions of the world's oceans, where plastics <5 mm in size have been found to be the most abundant type of marine debris (Browne et al., 2010; Eriksen et al., 2014). Eriksen et al. (2014) estimated surface water concentrations of microplastics of 0.2 per meter squared in open ocean waters of the GoM using data collected elsewhere between 2007 and 2013. This estimate is 2–3 orders of magnitude lower than our measurements on the intertidal shoreline of Mobile Bay, and it suggests

Table 1
Experimental results from a one-way ANOVA for both treatment effects and location effects (nested within treatments) on microplastics.

Microplastic	F	$DF_{(hyp,error)}$	$P_{treatment}$	F	$DF_{(hyp,error)}$	$P_{loc(trt)}$
Total	11.772	(1,14)	0.019*	1.809	(5,14)	0.176
Length	0.304	(1,14)	0.605	1.313	(5,14)	0.314
Mass	0.627	(1,14)	0.464	1.514	(5,14)	0.248

that microplastics accumulate on shorelines at higher concentrations in comparison with open waters. It is important to note that these samples were collected during the summer, depositional beach period and as such may have larger concentrations of microplastics than a winter, erosional beach.

Microplastic abundance did not vary among locations within the more marine-influenced or less marine-influenced groups, but it did vary significantly between the two groups (Table 1, Fig. 3). Indeed, values of microplastic abundance ranged from a minimum of 5 at site 2 to a maximum of 117 pieces per square meter at site 7 (Figs. 1, 3). These locations are approximately 35 km apart straight line distance and are both composed of sandy sediments, with the main difference between the two locations being a higher marine influence (i.e. higher salinity and exposure to tidal circulation) in the latter location. Site 2 is positioned on the western shore of Mobile Bay just south of the mouth of Dog River in an area that while exposed to daily tidal cycles experiences stratification and retains a freshwater surface layer, whereas site 7 is located on the southern side of a barrier island that is continuously exposed to ocean currents and tides. Overall the more marine-influenced locations had 50.6 ± 9.96 microplastics per m^2 (mean ± SE), and the more freshwater-dominated locations had 13.2 ± 2.96 microplastics per m^2.

Browne et al. (2010) found that in Plymouth Bay, England more microplastics were found at downwind sites in the estuary. Unfortunately, this does not hold true in Mobile Bay where in the spring and summer months predominate winds are out of the south (Kimball, 2009). Instead, we hypothesize that the differences found between these two groups may result from parallel differences in area and residence time of the input source. Mobile Bay has an area of approximately 1100 km^2 and water residence time from 3 to 160 days, whereas the much larger Gulf of Mexico has an area of 1.6 million km^2 and water residence time from 3 months to 250 years (Dinnel and Wiseman, 1986, Rivas et al., 2005). Therefore, plastic trash washed into the Gulf of Mexico would have much more time to degrade and break down into pieces smaller than 5 mm in comparison with trash in Mobile Bay, leading to larger pools of microplastics in the Gulf of Mexico in comparison with Mobile Bay (Kubota, 1994; Edyvane et al., 2004). Based on this, it can be expected to find a higher abundance of microplastics stranded on the shorelines of locations that are more exposed to tides, currents and winds from the Gulf of Mexico (i.e. sites 5 to 7 in this study) than from direct inputs from Mobile Bay (i.e. sites 1 to 4).

It is important to note that all microplastic pieces found during this study were secondary microplastics (i.e. resulting from the breakdown of larger plastics) and that no plastic manufacturing pellets were found (Figs. 4, 5). This further supports the idea that microplastic pollution in the northern Gulf of Mexico is a result of the degradation of plastic debris (Wessel, unpublished data).

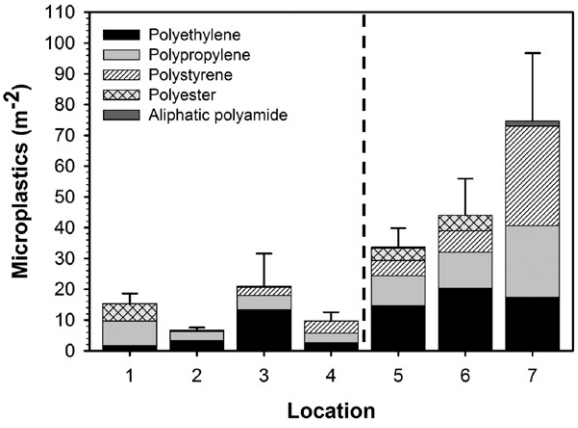

Fig. 3. The average ($n = 3$) abundance of total microplastics and abundance of each of the five polymers (polyethylene, polypropylene, polystyrene, polyester, and aliphatic polyamide) found as microplastics at the seven sampling locations. The most abundant polymers found were polyethylene (black) and polypropylene (gray). The dashed line separates the riverine locations (left) from the marine locations (right).

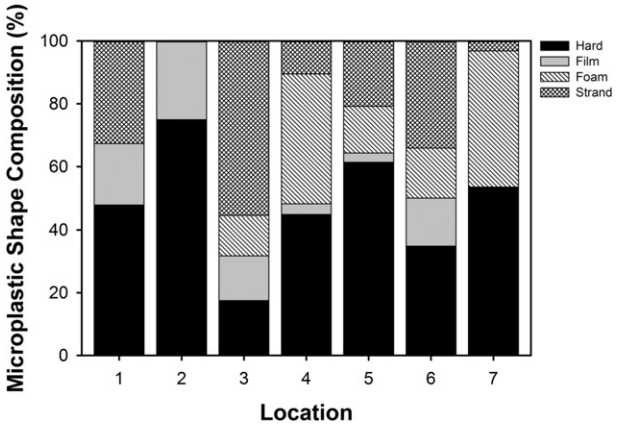

Fig. 4. The average microplastic shapes found at each location. Hard plastics (black bars) were the most abundant at 47.8%, followed by strands (hash lines) at 22.3%, foam (backslash) at 18.3%, and film (gray) at 12.2%.

The average (±SE) particle size and mass for all collected microplastics were 2.5 ± 0.48 mm and 4.0 ± 1.03 mg respectively, but this average is likely skewed towards the larger end since microplastics smaller than 0.2 mm were not collected for this study. Microplastic particle mass and size did not vary among locations within the more marine-influenced or less marine-influenced groups (Table 1, Fig. 6). Particle size did not vary between the two groups of locations, but particle mass was significantly ($F_{1,19} = 8.536$, $p = 0.009$) larger in the more marine-influenced (mean ± SE = 5.53 ± 0.97 mg) than in the less marine-influenced locations (2.92 ± 1.07 mg). While we don't know the exact density for each microplastic since we measured size and mass but not volume, this imbalance suggests microplastics deposited on locations that are more marine-influenced are denser overall than those deposited on less marine-influenced locations.

Despite the fact that only microplastics bigger than 0.2 mm were sampled we see that the size fraction from 0.2 to 1 mm makes up 38.6% of the 614 microplastics collected during this study. The other four size fractions (i.e. 1–2, 2–3, 3–4, 4–5 mm) each make up on average approximately 15% of the total. If this exponential trend continues with particles smaller than 0.2 mm making up even more of the total percentage it could have huge implications for impacting filter feeders in estuarine ecosystem which would be consuming this smaller size fraction. Due to this unknown any conclusions made by this study are limited to microplastics larger than 0.2 mm.

Five polymers were found in our samples: polyethylene, polypropylene, polystyrene, polyester and aliphatic polyamide (commonly called nylon; Fig. 3). All of the polymers were found more often at marine influenced locations than riverine locations and the differences inferred for particle density between more and less marine-influenced locations correspond well with differences in microplastic composition. More dense polymers (i.e. polystyrene, density = 0.45–1.04 g/cm^3; polyester, density = 0.96–1.37 g/cm^3; aliphatic polyamide, density = 1.15 g/cm^3) have lower prevalence than less dense polymers (i.e. polypropylene, density = 0.92 g/cm^3; polyethylene, density = 0.975 g/cm^3) in the area studied. However, the contribution by more dense polymers to the total microplastics sampled is generally higher in more marine-influenced than in less marine-influenced locations.

Polyethylene and polypropylene, the two most abundant polymers in this study, have many applications. Polypropylene is commonly used to produce single use items such as, bottle caps, food wrappers, and straws, while polyethylene is used for products such as, bottles, plastic bags, packaging, and fishing line. These results are consistent with the 2014 Alabama Coastal Clean-up (organized by the Ocean Conservancy and the Alabama Department of Natural Resources) data

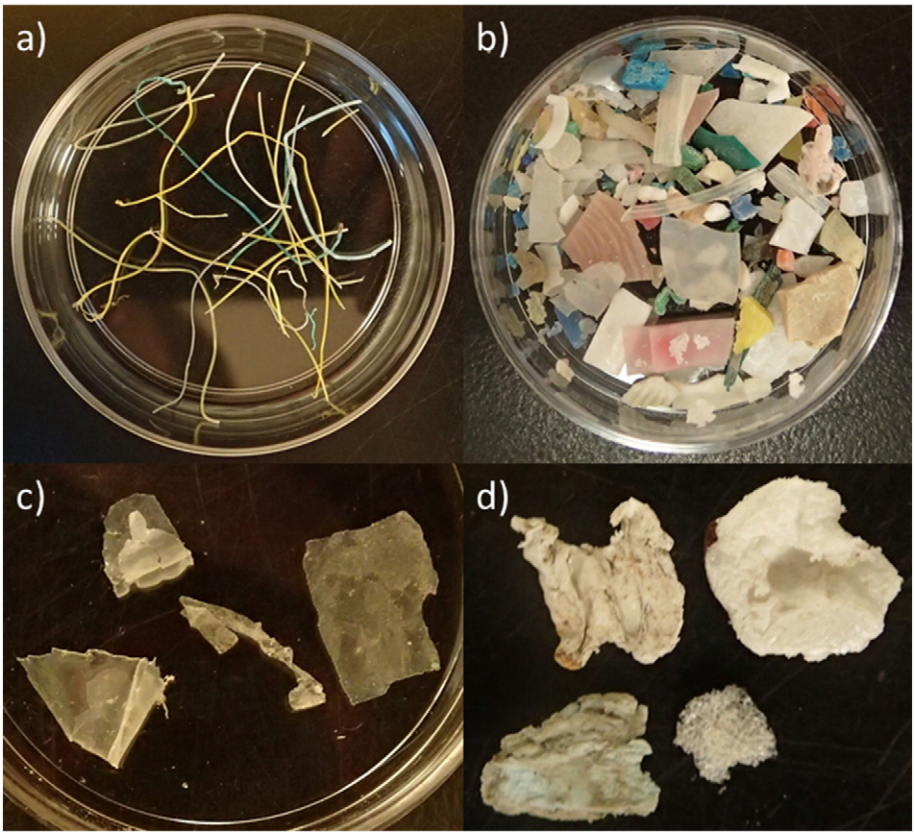

Fig. 5. Examples of the shapes of microplastics found in the GoM, a) strands, b) hard, c) film, d) foam.

which showed the six most abundant plastic marine litter items were (1) cigarette butts (2) bottle caps (3) food wrappers (4) beverage bottles (5) grocery bags and (6) plastic straws (ADCNR, unpublished data). The 29 sites included in this cleanup are all located along waterways in the two southernmost Alabama counties which are located on or drain into Mobile Bay; including Weeks Bay, Fort Morgan, and Dauphin Island (the three marine locations sampled) and Dog River and Daphne (two of the riverine locations sampled) further suggesting that these are secondary microplastics resulting from land-based debris sources (Sheavly and Register, 2007).

Our work provides new insights into the distribution, accumulation, sources and composition of microplastics. We show that microplastics are ubiquitous on the shorelines of Mobile Bay, a major estuarine system in the Northern Gulf of Mexico, and that shoreline microplastic concentrations are much higher than those predicted for open waters. The polymers polypropylene and polyethylene are predominant throughout the study area, whereas polystyrene, polyester and aliphatic polyamide are found in lower quantities. Interestingly, we find a stark difference throughout the bay related to microplastic composition and abundance; locations that are more directly exposed to marine current and tidal flow have larger microplastic abundance and diversity, and a higher contribution by the more dense polymers, in relation to locations that are less marine-influenced. The Gulf of Mexico is a vast reservoir, which may allow for sustained degradation time and development of large pools of microplastics. These results suggest that exposure to currents and tides from the Gulf may be a major determinant of microplastic accumulation on shorelines of the Gulf of Mexico. Areas leeward of currents and with strong tidal influence, where microplastics could accumulate in large quantities, may therefore be particularly critical and potentially damaging for organisms.

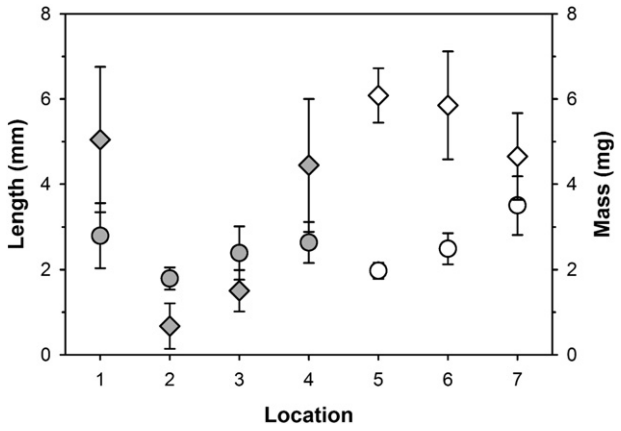

Fig. 6. The average ($n = 3$) microplastic particle size and mass for each location sampled. Size (measured as length, mm) are circles and masses (mg) are diamonds, standard error bars are included. Freshwater locations are shaded with gray and marine locations are in white.

Acknowledgments

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