

**Key Points:**

- The sources, behavior, fate, and effects of microplastics are inextricably tied to those of their plastic product and macrodebris precursors, as well as the nanoplastics into which they fragment
- Microplastic sampling and characterization methods are evolving, but their inadequacies still hamper efforts to evaluate the true extent of their presence and consequences in the built and natural environments
- Microplastics are a global/multimedia phenomenon; hence, they cannot be adequately understood, or related concerns resolved, in the context of the marine environment alone or a single discipline

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## A Global Perspective on Microplastics

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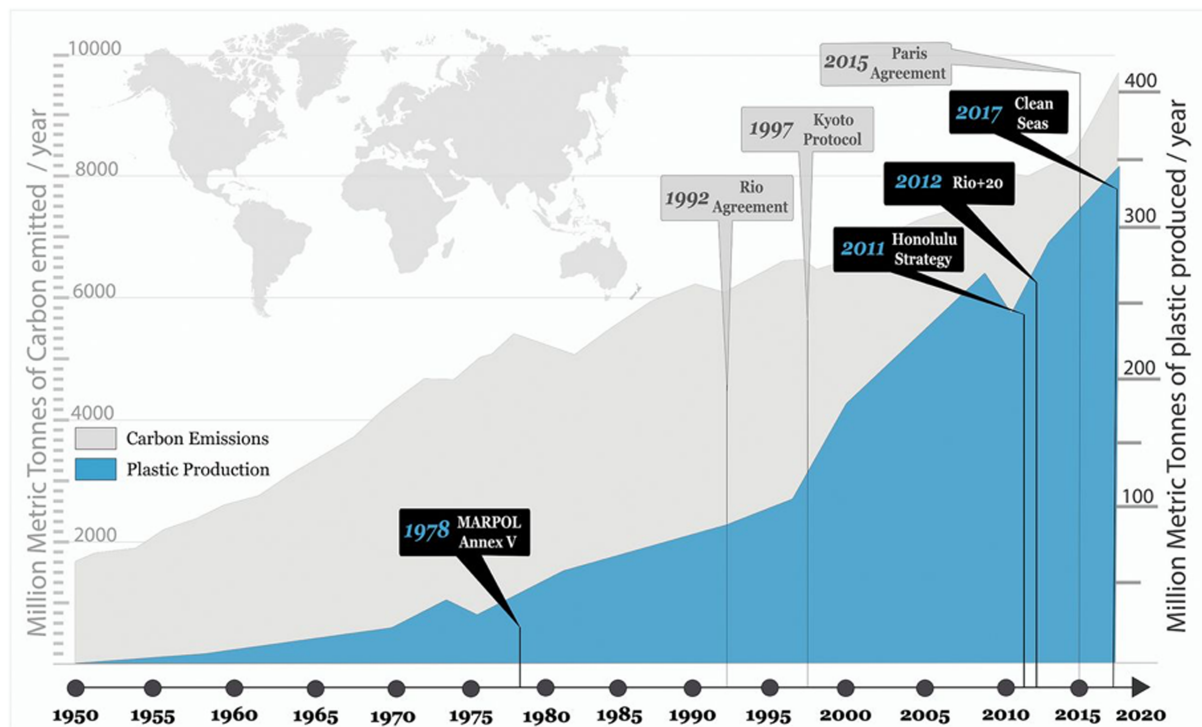
**Abstract** Society has become increasingly reliant on plastics since commercial production began in about 1950. Their versatility, stability, light weight, and low production costs have fueled global demand. Most plastics are initially used and discarded on land. Nonetheless, the amount of microplastics in some oceanic compartments is predicted to double by 2030. To solve this global problem, we must understand plastic composition, physical forms, uses, transport, and fragmentation into microplastics (and nanoplastics). Plastic debris/microplastics arise from land disposal, wastewater treatment, tire wear, paint failure, textile washing, and at-sea losses. Riverine and atmospheric transport, storm water, and disasters facilitate releases. In surface waters plastics/microplastics weather, biofoul, aggregate, and sink, are ingested by organisms and redistributed by currents. Ocean sediments are likely the ultimate destination. Plastics release additives, concentrate environmental contaminants, and serve as substrates for biofilms, including exotic and pathogenic species. Microplastic abundance increases as fragment size decreases, as does the proportion of organisms capable of ingesting them. Particles <20  $\mu\text{m}$  may penetrate cell membranes, exacerbating risks. Exposure can compromise feeding, metabolic processes, reproduction, and behavior. But more investigation is required to draw definitive conclusions. Human ingestion of contaminated seafood and water is a concern. Microplastics indoors present yet uncharacterized risks, magnified by the time we spend inside (>90%) and the abundance of polymeric products therein. Scientific challenges include improving microplastic sampling and characterization approaches, understanding long-term behavior, additive bioavailability, and organismal and ecosystem health risks. Solutions include improving globally based pollution prevention, developing degradable polymers and additives, and reducing consumption/expanding plastic reuse.

### 1. Overview

Microplastics (1 to 5,000  $\mu\text{m}$  particles) captured widespread attention after reports detailed massive “garbage patches” in the world’s great oceanic gyres. Concern followed over possible negative impacts on marine life. While plastic wastes in the environment is truly a global and multi-media issue, popular press and scientific attention has predominantly been “ocean-centric” (e.g. Cole et al., 2011). Like climate change and persistent organic pollutants, plastic debris exemplifies our capacity to alter the environment on a global scale. Villarrubia-Gómez et al. (2018) argued that marine plastic contamination is irreversible and globally ubiquitous and therefore meets two of the three conditions for a chemical pollution planetary boundary threat. The third condition is demonstrated widespread ecological disruption. Investigation of the possibility for such disruption has only recently begun but will become more critical as plastic contamination rises. Indeed, Koelmans et al. (2017) recently argued that it is time to move beyond conjecture and proposed a frame work for evaluating toxicological risks of microplastics.

Microplastics are found in diverse forms, including spheres, fragments, and fibers. Most (with the exception of intentionally manufactured microbeads) arise from the deterioration of larger plastics (macroplastics). Microplastics fragment into ever-smaller debris over time, eventually becoming nanoplastics (<1  $\mu\text{m}$ ; Lambert & Wagner, 2016; Hartmann et al., 2019). Hence, microplastics are largely a transitional state between macrodebris and nanomaterials. Besseling et al. (2018) estimated that fragmentation of spherical microplastics could generate >10<sup>14</sup> times greater numbers of nanoparticles. To understand microplastic sources, fate, and consequences, one must consider the continuum, from plastic products/debris to microplastics and nanoplastics. Hence, this review will encompass all three size classes.

Despite growing attention, the actual amounts of plastics in environmental compartments (terrestrial, marine, freshwater, and atmospheric) and their ecological significance are still unclear. This is in part due to the recency of attention and lack of adequate sampling and analysis approaches, as well as the immensity and



**Figure 1.** While the global emission of carbon exceeds that of plastic production, the rate of increase of the latter now exceeds that of the former. Figure from Borrelle et al. (2017).

diversity of the oceans. Estimates of the amount of floating plastic in the ocean are as high as 236,000 metric tons (Van Sebille et al., 2015). Jambeck et al. (2015) suggested that 4.8 to 12.7 million tons of plastic debris per year enter the ocean and projected cumulative inputs will increase tenfold by 2025.

Most published plastic debris surveys have focused on the ocean's surface (e.g., C  zar et al., 2014), but efforts are now expanding into deeper waters, sediments, freshwaters, soils, air, and biological systems. In part, this is based on ease of sampling and a predilection toward floating plastics. This is being remedied as the research field matures. Microplastics have recently been discovered in presumably pristine locales, including Arctic sea ice (Peeken et al., 2018), the Antarctic (Waller et al., 2017), remote mountain ranges (Allen et al., 2019), and deep ocean trenches (Jamieson et al., 2019). As is common with emerging environmental issues, much early work focused on describing the extent of microplastic contamination, although sampling and analytical methods are not yet up to the challenge. Research is now also expanding to the processes of microplastic formation, transport, fate, organismal exposure, and ecosystem effects.

As immense as the issue of microplastics in the world's ocean is, it is only part of a larger, more complex reality. Most waste plastics are disposed of in landfills, incinerated, or recycled, although much is mismanaged and enters the natural environment. Over the last 70 years the nations of the world have become increasingly dependent on plastics. Between 1950 and 2015 the annual growth rate of production has been 8.4% (Geyer et al., 2017). Emerging and poor countries are now adopting usage of plastics *en masse* (Lwanga et al., 2017), resulting in an upsurge in global plastic manufacture and consumption. The rate of plastic production has recently surpassed that for carbon emissions (Figure 1). Ironically, affluent nations have shipped substantial amounts of plastic wastes, including obsolete electronics (*e-waste*), to poorer countries for recycling. In many cases these materials have been mishandled and much of the remnants (after removal of valuable components such as copper and circuit boards) discarded improperly or even burned (Asante et al., 2016). For these reasons and a lack of waste management infrastructure, the Asian Pacific region is believed to have overtaken Western countries as the major contributor of plastic debris to coastal ocean waters (Jambeck et al., 2015). However, China, a major waste recipient, recently disrupted the plastics recycling market by restricting foreign imports (Tan et al., 2018). Other importing nations are following suit. This may alter future plastic debris distributions and force more affluent countries to invest in solutions.



To comprehend and ultimately resolve the microplastics problem, we must consider the nature, usage, and fate of plastics in terrestrial, atmospheric, and aquatic compartments. We must balance the positive and negative consequences of plastics, in order to prioritize actions to best protect global health. Currently, most people researching microplastics in the environment have operated largely within individual specialties, for example, polymer chemistry, waste management, atmospheric, terrestrial, freshwater, and marine science. Delineation and resolution of the plastics issue will require wider participation/collaboration, adoption of more holistic, interdisciplinary approaches, and recognition of plastic pollution as a risk to the global environment. This review is intended to facilitate that transition and will conclude with the identification of key challenges to achieve these ends.

## 2. The Nature of Plastics and Microplastics

While the public often assumes that all plastics are compositionally the same and thus behave analogously in the environment, this is not the case. To understand the behavior, fate, and consequences of microplastics, we must first consider their composition and diversity. Identification of microplastics in the environment presents a multidimensional challenge that is yet unmet. Their complexity parallels that of naturally occurring, particulate organic matter (Hoellein et al., 2019). Plastics (and thus microplastics) vary in chemical composition, physical form, size, texture, and shape. These characteristics evolve while in use and after discard.

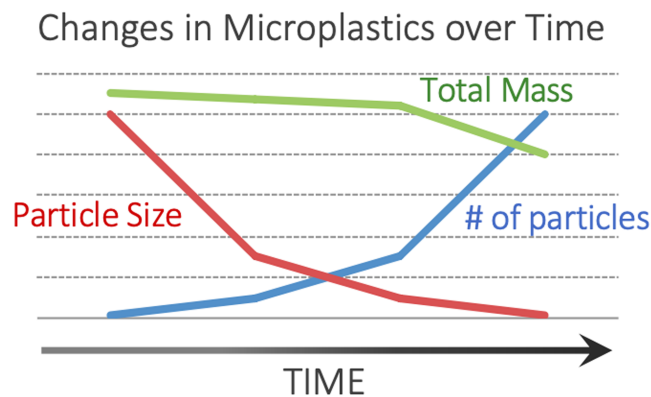
Intentionally manufactured microplastics, designated *primary* microplastics, include microbeads in personal care products and industrial abrasives for delicate surfaces. Microbeads are also used in cleaning agents, coatings and paints, drilling fluids in the oil and gas industry, and as precursor resins and pellets for the manufacture of finished plastic products. In the U.S. the *Microbeads Free Waters Act of 2015* was enacted to eliminate microbeads from rinse-off personal care products (McDevitt et al., 2017), but not from nonrinse off (e.g., sunscreen and cosmetic makeup) or industrial applications. Similar regulations have been enacted elsewhere. *Secondary* microplastics are formed from the fragmentation of larger plastics during usage (e.g., wear particles from tires) or after disposal. Secondary are far more abundant than primary microplastics. Environmental half-lives of plastics vary by polymer type and ambient conditions but range from days to centuries (Ward et al., 2019). Most have not been evaluated scientifically. Andrady (2017) postulated that a majority of plastic fragmentation occurs on land due to greater ambient temperatures, frictional forces, and UV exposure.

### 2.1. Uses and Properties of Plastics

Due to their attributes, synthetic polymers have supplanted many naturally derived materials in modern society (Lebreton & Andrady, 2019). Applications include single-use food and beverage containers, thermal insulation, home and workplace furnishings, electrical and electronic devices, vehicle interiors, toys, fabrics, surface coatings, and even medical devices (e.g., artificial joints, incubators, intravenous (IV) fluid bags, and drug delivery devices).

The design of plastics determines their properties and fitness for desired applications. Plastics are composites of long chain organic polymers. Finished products may be homogeneous in terms of constituent polymer or contain different types blended or cross-reacted to achieve the desired characteristics. Polymer chains are produced by combining chemical monomers, often derived from fossil fuels, into strands of repeating units. Polymers also occur naturally in molecules such as biodegradable deoxyribonucleic acid or starch, as well as more environmentally persistent cellulose and chitin. Microorganisms capable of degrading these natural polymers have evolved over time. In contrast, synthetic polymers have only been produced in large amounts since about 1950. For this reason and their compositional features, most synthetic polymers exhibit greater resistance to biodegradation.

The densities of plastics vary by composition and span those of water. Most early observations of plastics focused on macrodebris floating at the water's surface or stranded on shorelines. These include low-density polymers used in single-use containers such as polyethylene and polypropylene. However, many other polymers (e.g., polyethylene terephthalate, polycarbonate, and polyvinyl chloride) are denser than water and thus are expected to sink. In aquatic environments, even these simple predictions as to fate may be misleading, as over time most surfaces develop a biofilm (Zettler et al., 2013) or form aggregations, which may eventually cause even buoyant plastics to sink. Polystyrene foam is common in fishing floats but will sink after air



**Figure 2.** Proposed relationship between microplastic size, particle number, and total mass over time. We postulate that as particle size decreases and surface area increases, environmental reactivity will increase. This may result in a decrease in total mass due to enhanced biodegradation.

spaces within become waterlogged. In contrast, surface tension can maintain dense plastic resins at the water's surface.

Organic or inorganic additives are used to modify plastic properties such as color, flexibility, flame retardancy, and ultraviolet (UV) resistance. These additives may be present at percent by weight levels and will travel with fragments formed from the parent products. Nonpolymeric fillers, either functional or simple extenders, may also be added to plastics during their formation to modify performance or reduce cost. It is noteworthy that these amendments can even alter the overall density of the product and thus its physical fate in aquatic systems. Additives, unreacted monomers, degradates, and other materials (including nanoparticles) may be released from the polymer matrix as a function of their properties and environmental conditions. Indeed, researchers reported that some of these plastic-associated components may contribute to oceanic dissolved organic carbon (DOC) pools (Romera-Castillo et al., 2018).

A plastic product's mode of use and disposal influences its fate. To illustrate, automobile tires are subject to frictional road wear and weathering

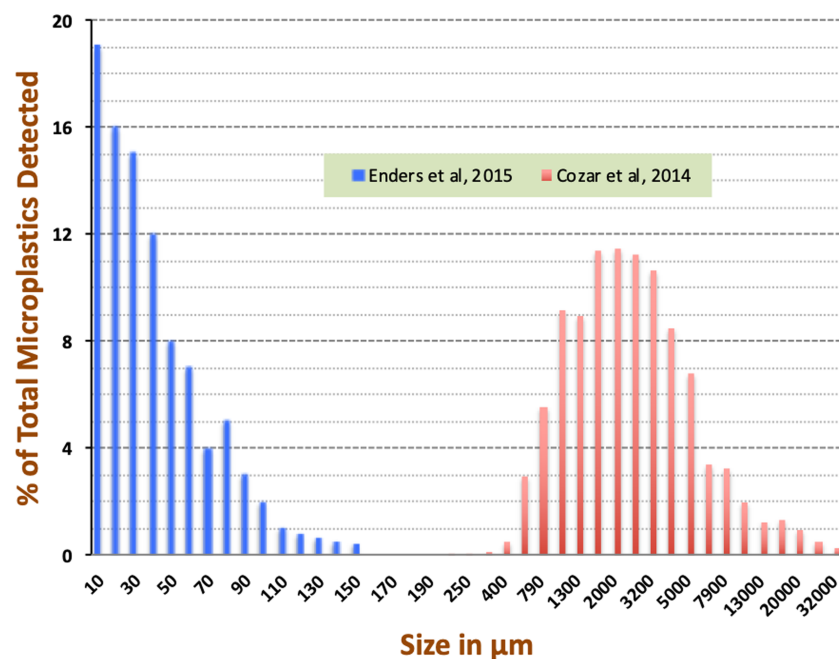
while in use. In contrast, the plastic casings of electronics typically reside indoors and release minimal microplastics during their in-use lifetimes. Diversity of composition and the presence of potentially toxic additives in plastics complicate reuse and environmental consequences. For example, if the host polymer breaks down rapidly in the environment, the additives therein may be released. In recycling, banned additives may carry over into the new products. For example, flame retardant polybrominated diphenyl ether (PBDE) polymer additives were removed from production in the U.S. in 2004. However, residues may be present in contemporary carpet underlayment formed from recycled materials (DiGangi et al., 2011). Recycling of e-wastes in developing countries often involves shredding and burning plastics and hence releases microplastics locally (Labunska et al., 2013).

## 2.2. Weathering and Degradation of Plastics

Plastics are vulnerable to weathering to varying degrees. Chemical oxidation of the polymer as a result of exposure to sunlight is often the most impactful (Andrady, 2015). As noted above, additives may reduce such degradation. Photooxidation is most rapid at the water surface, on beaches, and in exposed terrestrial scenarios, negligible if shielded in aquatic sediments, soil, or landfills. Weinstein et al. (2016) noted that biofilm formation on plastic surfaces reduced UV light penetration by up to 99%. Interestingly, Khaled et al. (2018) reported that incorporation of brominated flame retardant additives into polystyrene film increased UV absorption and subsequent photooxidation of the polymer. They observed that byproducts were generated from the flame retardants themselves and from the reaction of the polystyrene with bromine radicals. The amount of degradation products leached into water constituted up to 14% of the weight of the original polymeric film. Weathering of plastics by UV oxidation may also increase vulnerability to later biodegradation.

Some polymers are composed of monomers derived from renewable, nonpetroleum sources, for example, rayon and cellulose acetate (used in textiles and cigarette filters). Hartmann et al. (2019) included these as "plastics," although others have excluded them due to their cellulose-derived origin. Additionally, polyhydroxyalkanoate from bacterial precursors and polylactic acid from plant starch have recently been produced to be more inherently biodegradable (Harrison et al., 2018). The goal is to allow complete degradation to CO<sub>2</sub> after the end of product service life, as partial breakdown may generate intermediates of unknown properties, as well as microplastics. The rate of polymer biodegradation increases as particle size decreases and surface area increases (Chinaglia et al., 2018), although this has not been well investigated under marine conditions. This may be an important factor in long-term fate of microplastics (Figure 2). Characteristics of the surrounding environment are also controlling (Dilkes-Hoffman et al., 2019). For example, the above biopolymers are denser than water and hence will sink. There, it may encounter lower ambient oxygen levels, temperature, and light exposure, reducing the subsequent rate of degradation.

Recent work indicates that certain microorganisms may be capable of degrading petrobased synthetic polymers. For example, Yang et al. (2015) observed that the gut bacteria of mealworms can slowly breakdown



**Figure 3.** Microplastic size distributions depend on sampling location (geographically and vertically in the water column) and analytical methods applied. For example, Enders et al. (2015) pumped North Atlantic surface waters through a 10  $\mu\text{m}$  filter and then analyzed microplastics with Raman microspectroscopy. C  zar et al. (2014) collected floating microplastics with a 200  $\mu\text{m}$  net during the Malaspina global circumnavigation. They selected candidate microplastics with a dissecting scope and evaluated composition of random particles with Raman spectroscopy. Selection of surface waters, use of large mesh nets ( $>200 \mu\text{m}$ ), and FTIR detection (minimum detection limit 20  $\mu\text{m}$ , but often larger) has been common and hence will underestimate the contribution of small microplastics or dense polymers that sink.

polystyrene, although residues remain. Yoshida et al. (2016) reported that bacteria exposed to polyethylene terephthalate at a recycling site produced enzymes that could degrade it to its constituent monomers. However, polymers with greater crystallinity, such as polyethylene, may be more resistant. Nonetheless, Brandon et al. (2018) observed similar rates of degradation of polyethylene and polystyrene by mealworms and that this was facilitated by adaption of their microbiomes. Microbial growth during use is a concern in a number of plastic applications, such as kitchenware. Thus, some manufacturers have added antimicrobial agents, for example, triclosan, to products. Alternatively, the so-called oxodegradable plastics are imbued with additives (e.g., transition metals) to accelerate polymer oxidation and fragmentation of the plastic (Ammala et al., 2011). This was initially promoted as an attractive feature for plastic sheeting (also known as plastic mulch or plasticulture) used to block weeds and maintain soil moisture and temperature in crop farming (Steinmetz et al., 2016). However, biodegradation of the residual plastic has been found to be slow and to release microplastics. Hence, the European Union has taken steps to restrict their usage (European Union, 2018). However, a new polymer called poly (diketoenamine) was recently synthesized that may be readily disassembled into its monomers (Christensen et al., 2019). This allows flexibility of reuse and an opportunity to separate out unwanted additives from previous applications.

### 2.3. Analysis of Microplastics

An overview of current sampling and analytical approaches for determining microplastics in complex environmental media is provided here to allow the reader to evaluate the completeness and accuracy of the available literature detailing their presence in the global environment. The first crucial step for quantifying microplastics in matrices from the field is collecting a representative sample consistent with the desired research objectives. Many published studies have focused on specific zones where plastics are obvious, such as the sea surface, shorelines, and beaches. Water sampling has often been conducted by towing a mesh net (often 300  $\mu\text{m}$ ). However, this distorts the true size distribution of microplastics in the water, as shown in Figure 3, which compares data from C  zar et al. (2014) versus Enders et al. (2015). Further, Chae et al.

(2015) collected orders of magnitude greater microplastics from water when using a 20  $\mu\text{m}$  hand net versus 300  $\mu\text{m}$  trawl net. In addition, higher-density and biofouled microplastics will sink; thus, water surface-focused approaches will not collect these.

Organisms may be evaluated for microplastic content in their entirety, or specific organs targeted. In any event, researchers (e.g., Erni-Cassola et al., 2017) commonly utilize a digestion step to eliminate labile tissues, leaving behind the more recalcitrant microplastics. Evaluation of small microplastics (on the order of 10  $\mu\text{m}$ ) and nanoplastics that may have penetrated cell membranes should be considered as these may have greater toxicological implications (Triebkorn et al., 2018) than those in the lumen of the gut (which may be quickly egested). Care to prevent and monitor contamination by nonsample-related microplastics during collection, processing, and detection are essential. Lab environments, where sample processing occurs, are major sources of microplastics due to the abundance of synthetic fibers and other plastic products indoors. This portends the extent of our personal exposure to microplastics indoors.

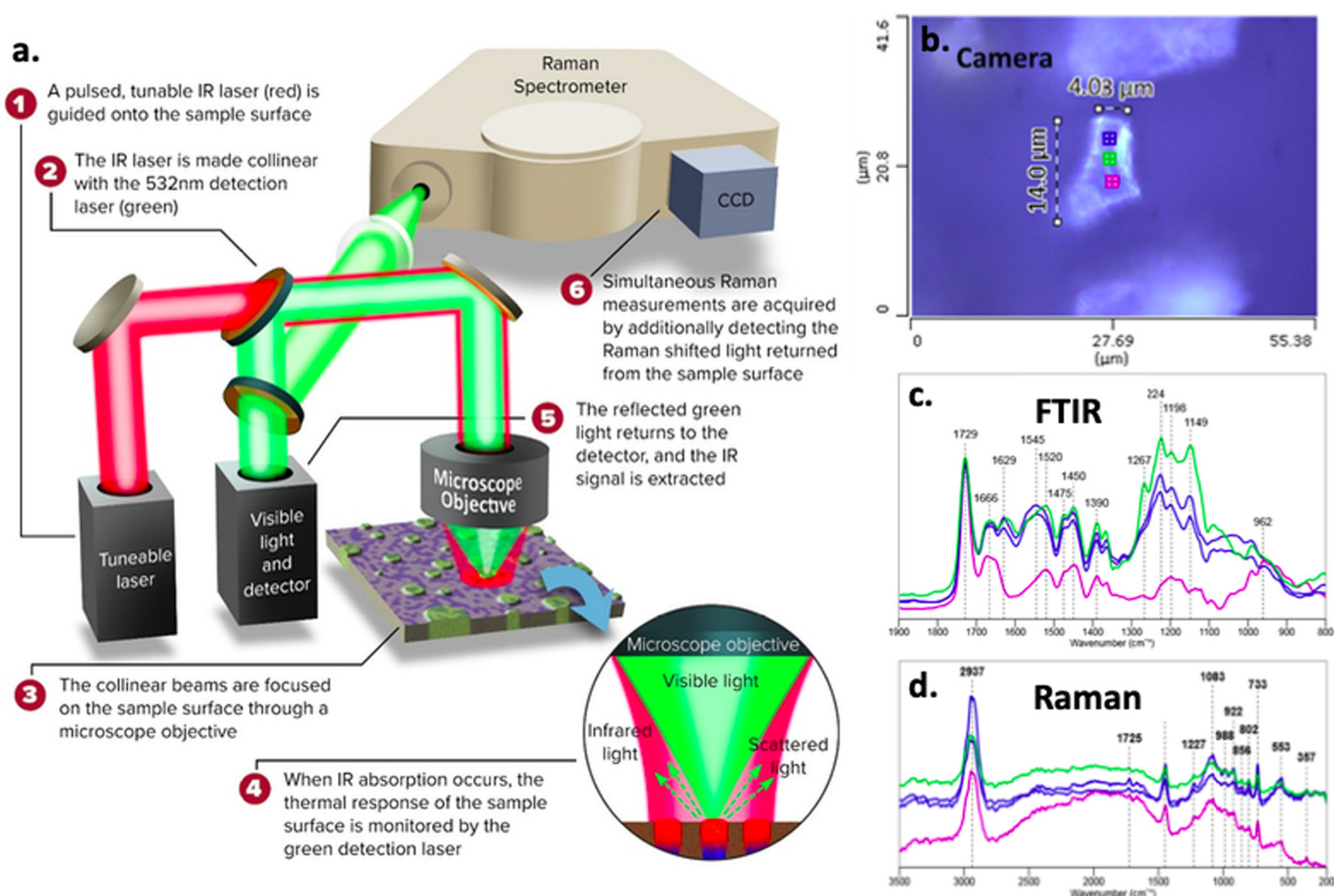
To understand the fate and consequences of microplastics in the environment, we must be able to measure their full range of sizes and composition. Efforts to date examining their distribution and abundances have been hampered by inadequate methods. Matrices where microplastics reside include water, sediment, biological tissues, soil, wastewater sludge, and air. Approaches to sampling, cleanup, and detection of microplastics in these media are still evolving. Multistep procedures are common in environmental chemistry due to the need to extract the analytes from the bulk matrix and remove interfering materials before final microplastic detection and quantitation. Ideally, the detection technique provides both quantitative and qualitative information (e.g., polymer type and quantity, additive content, fragment dimensions, and shape). Analysis should be automated where feasible to reduce costs and accelerate sample throughput. To date, researchers in the field have utilized different techniques for extraction from environmental matrices (Fuller & Gautam, 2016; Hurley, Lusher, et al., 2018; Hurley, Woodward, et al., 2018; Wagner et al., 2017) and for polymer identification (Fries et al., 2013; K  ppler et al., 2018; Mintenig et al., 2017). Differences in method effectiveness make comparison of data between studies more difficult.

As microplastics breakdown continuously in the environment, smaller fragments become increasingly abundant over time (see Figures 2 and 3). While the fate of these minute particles has not yet been determined, they hold the key to the ultimate destiny of microplastics in the environment. Nonetheless, the majority of published reports have focused on materials >100  $\mu\text{m}$  due to methodological constraints. Published studies also present the presence of microplastics in different ways, for example, number or weight of microplastics per sample volume or weight. This further impairs comparison of studies.

Several reviews of microplastic detection methods have been published (e.g., Mai et al., 2018; Prata, 2018; Zarfl, 2019). Early approaches often relied on light microscopy. However, this technique cannot provide chemical composition and thus is weak for differentiating synthetic polymers from sample interferences. Fourier transform infrared spectroscopy (FTIR) has been widely used for polymer identification, producing spectra that can be matched to library standards. The development of FTIR microspectroscopy, allowing particle chemical mapping, has substantially advanced microplastic characterization capabilities. In sophisticated instruments sample filtrates may be automatically scanned for microparticles and fibers and tentative polymer types assigned using, for example, a focal plane array detector (e.g., Primpke et al., 2017). Unfortunately, conventional FTIR is typically limited to targets >10  $\mu\text{m}$  due to diffraction limit considerations. Raman microspectroscopy can provide detection down to about 1  $\mu\text{m}$  and is being increasingly utilized (Schymanski et al., 2018). Polymers may be weathered and thus produce spectra deviating from virgin standards, complicating identification. In response, some researchers have created their own libraries of weathered materials to assist identification (e.g., Choy et al., 2019). Attenuated total reflectance FTIR and atomic force microscopy have also been used but require physical contact of the probe with the targeted material. Thus, these tools may not be optimal for samples containing numerous minute targets. Optical photothermal infrared spectroscopy, offering submicron spatial resolution, has recently been commercialized. This technique uses a visible light probe to measure the photothermal response of targeted particles following IR absorption (Figure 4). Configurations are available that permit both IR and Raman scans to be performed on a given target, providing increased compositional information (Marcott et al., 2019).

Alternatively, Erni-Cassola et al. (2017) applied a lipophilic fluorescent dye (Nile red), in combination with fluorescence microscopy, to reveal microplastics in samples. Use of image analysis software allowed





**Figure 4.** Schematic of an optical photothermal IR/Raman spectrometer (a), a visible image of a microparticle in an indoor dust sample (b), and corresponding FTIR (c) and Raman (d) spectra. The microparticle was tentatively identified as a poly (methyl methacrylate) based on comparison to library spectra. Analysis and graphics courtesy of Debra Cook and Jay Anderson, Photothermal Spectroscopy Corp., Santa Barbara, CA, USA.

automated measurements. Prior to detection, the authors applied a hydrogen peroxide treatment (see discussion below) to reduce/eliminate fluorescent biogenic materials. However, some less intense fluorescence by residual chitin was observed. Fluorescence intensity was also less for more polar polymers (e.g., polycarbonate, polyurethane, polyethylene terephthalate, and polyvinyl chloride) than lipophilic polymers such as polyethylene and polypropylene. Fluorescence may also be used to prescreen targets for further micro FTIR or Raman investigation (e.g., Maes et al., 2017). Finally, destructive techniques such as pyrolysis-GC/MS can be employed, whereby microplastic(s) is vaporized at between 600 and 700 °C and resulting components chromatographically separated and identified by mass spectrometry (Käppler et al., 2018). However, destructive techniques such as this do not allow evaluation of particle shape and size.

Alternatively, samples can be preserved to isolate the desired size ranges. Resulting fractions may then be extracted/dissolved and then subjected to pyrolysis or other techniques. For example, Ceccarini et al. (2018) applied differential solvent extraction, pyrolysis gas chromatography/mass spectrometry (GC/MS), FTIR, size exclusion chromatography, and nuclear magnetic resonance to the analysis of microplastics in beach sediment samples. They observed (1) substantially greater amounts of microplastics than commonly reported in samples due to their detection of microplastics less than 20  $\mu\text{m}$ ; (2) extensive oxidation of olefinic and polystyrene polymers due to weathering; and (3) fractionation of different microplastics by size, polymeric composition, and degree of chemical weathering across beach transects. They further observed that denser microplastics (such as polyester, polyamide, and polyvinyl chloride), as well as oxidized microplastics, accumulated preferentially in aquatic versus beach sediments.

Environmental samples commonly contain interfering, nonplastic materials. Widely used treatments employ the low specific gravity and high chemical resistance of synthetic polymers (Stock et al., 2019) to remove interfering materials. Flotation/sedimentation to separate microplastics from denser materials, such as minerals, is regularly utilized. This may be conducted in simple glass separatory funnels or more elaborate apparatuses, such as the stainless steel Munich Plastic Sediment Separator (Coppock et al., 2017). As some polymers (e.g., polyethylene terephthalate, acrylonitrile butadiene styrene, and nylon) have densities exceeding water, a high-density saline solution (e.g., NaCl, NaI, or  $\text{ZnCl}_2$ ) may be employed to facilitate their flotation. In addition, researchers often employ a step to destroy interfering natural materials such as biofouling on plastic surfaces and cellulose and chitin particles (Stock et al., 2019). Hydrogen peroxide and Fenton's reagent are most commonly employed, although alternatives include acid, base, or a suite of enzymes that target specific classes. Care must be exercised to include the full range of polymers during density separation and not destroy more labile polymers during caustic treatment. Differential extraction may also be used. For example, Fuller and Gautam (2016) applied a two-step, pressurized fluid extraction with different organic solvents to separate microplastics from municipal waste and industrial soil samples. The first step used methanol at 100 °C to remove coincident fats and oils. The second step employed methylene chloride at 180 °C to yield the plastics. The extraction process could be conducted using an automated commercial instrument. The extracted material solidified after solvent evaporation into a composite plastic residue. Limitations were that shape and size data for individual particles were lost, and interpretation of the combined IR spectra from multiple, coincident polymers was more difficult. Alternatively, C. Liu, Li, et al. (2019) quantified polycarbonate and polyethylene terephthalate microplastics in a variety of matrices by first depolymerizing them via alkali-assisted thermal hydrolysis. They then used liquid chromatography/mass spectrometry (LC/MS) to detect the resulting bisphenol A and p-phthalic acid, respectively.

In summary, available sampling and analysis approaches have rapidly advanced in the past decade, yet they still fail to fully meet the challenges presented by microplastics in the environment and must be improved. Data on environmental microplastics levels should be scrutinized for methodological rigor and completeness. In most studies to date, published concentration values are underestimates, as they do not encompass important environmental compartments, polymer types, microplastic or nanoplastic sizes, or chemically weathered materials.

### 3. Microplastics in the Indoor and Terrestrial Environment

Residents of developed countries spend >90% of their lives indoors (Bernstein et al., 2008), and homes and workplaces are increasingly airtight and insulated with additive-treated insulation such as polystyrene. Consequently, our exposure to microplastics from inhalation and dust ingestion indoors may have toxicological consequences, but scant research exists on the subject. In one of the few published studies, Dris et al. (2017) reported indoor air concentrations of microfibers of between 1.0 and 60.0 fibers  $\text{m}^{-3}$ , exceeding outdoor levels (0.3 to 1.5 fibers  $\text{m}^{-3}$ ). Indoor microfibers consisted of 67% natural or hybrid materials (primarily cellulose fibers, acetate cellulose, or keratinous wool). The remaining fibers were wholly synthetic polymers, dominated by polypropylene (Dris et al., 2016).

Polymer-based products often contain chemical additives (e.g., flame-retardants, dyes, plasticizers, and UV-inhibitors). Such additives were long-assumed to be completely sequestered within the polymer matrix. However, we now recognize that they may enter indoor air and dust by volatilization, polymer degradation, and fragment dispersal. Additives in indoor dust samples have been shown to correlate with the number of polymer-related consumer products in the room (Marklund et al., 2003). A review by Lucattini et al. (2018) described multiple studies detecting polymer additives in indoor air and dust. Indoor levels generally exceed those outdoors (Melymuk et al., 2016; Wong et al., 2018). Potential health effects may be linked. For example, Pauly et al. (1998) examined 114 human malignant lung specimens and found that 87% contained cellosic or other polymeric fibers.

Volatilization of chemicals (e.g., styrene, vinyl chloride, and formaldehyde) from polymers is a major source of indoor air pollution; however, microplastic dispersal is an underappreciated pathway for semivolatile chemical dissemination. Concentration variations in indoor dust samples of up to 6 orders of magnitude for phthalates and flame retardant additives have been reported (Harrad et al., 2008; Lucattini et al., 2018); likely due to the heterogeneous distribution of microplastics within the indoor spaces sampled. Webster

et al. (2009) supported the premise that additive-laden microplastics play a role in contaminant dispersal. Using X-ray energy dispersive spectrometry, they imaged bromine-rich sites (a marker of brominated flame retardant additives) on microparticles in indoor dust samples from homes and vehicles. The particles themselves consisted of a calcium-containing acrylic polymer (calcium carbonate is a common polymer filler; Thenepalli et al., 2015).

Suzuki et al. (2009) detected microplastics in indoor dust samples from Japanese households and work places. Again, bromine-rich clusters within the dust samples were observed. Particle textures, colors, and fibrous shapes were consistent with microplastics and textile fibers, ranging in size from 500 to 1,500  $\mu\text{m}$ . Wagner et al. (2013) identified BDE-209 (a common flame retardant) containing microplastics in dust using Raman microspectroscopy. Fluorescent pigments, dyes, and optical brightening or whitening agents are also widely used in the plastic and textile industry (Christie, 1994). Dehghani et al. (2017) employed fluorescence microscopy to assess microplastics in urban street sweeping samples from the central district of Tehran, Iran. Fluorescent particles and fibers were visible in all samples. Samples were also analyzed by energy dispersive X-ray spectroscopy for Al, Na, Ca, Mg, and Si, elements common in polymer antioxidants. Concentrations of microplastics in the dust samples ranged from 2.8 to 20 particles  $\text{g}^{-1}$  and from <100 to 5,000  $\mu\text{m}$  in size. C. Liu, Li, et al. (2019), using the thermal hydrolysis/LC MS approach previously described, reported polyethylene terephthalate levels in Chinese indoor and outdoor dust up to 120,000 and 9,020 mg/kg, respectively. Polycarbonate concentrations were lower, that is, 4.6 and 2.0 mg/kg, respectively. The authors also employed FTIR microscopy and confirmed substantial numbers of polyester fibers in the dust, supporting the polyethylene terephthalate findings. The authors projected that the estimated daily intake of polyethylene terephthalate via dust ingestion for infants was 89,700 ng/kg-bw/day (C. Liu, Li, et al., 2019).

Catarino et al. (2018) compared microplastic exposure from eating mussels to that acquired from indoor dust fallout during the meal. The authors concluded that the latter likely contributed orders of magnitude more microplastics than did the former, highlighting the importance of human exposure in the indoor environment.

### 3.1. Terrestrial Inputs of Microplastics to the Oceans

Plastics are manufactured, used, and predominantly first disposed of on, or into, soils. Plastics at the soil surface are subjected to greater UV exposure, abrasion, and temperatures than water-immersed materials (Ng et al., 2018). Upon entering soils, microplastics can penetrate vertically via water infiltration (O'Connor et al., 2019), facilitated by wet/dry weather cycles or tilling (Rillig, Ingraffia, et al., 2017) and by the actions of soil organisms (Rillig, Ziersch, et al., 2017). Smaller particles are more likely to move horizontally and vertically. Panno et al. (2019) recently detected microplastics, exclusively fibers, up to 15.2 particles  $\text{L}^{-1}$  in 16 of 17 groundwater samples from two karst aquifers in Illinois (USA). They hypothesized these microplastics may have been derived from drainage from private septic systems. Treated wastewater effluents are increasingly being injected into aquifers to replenish groundwater. Oil and gas production water and other industrial wastes may contain microplastics and are also injected for disposal purposes. As to organismal uptake and transfer, Lwanga et al. (2017) measured microplastic and macroplastics from discarded packaging in home gardens in southeast Mexico. Analyses showed that microplastics increased approximately tenfold between soil (0.87 particles  $\text{g}^{-1}$ ), earthworms (14.8 particles  $\text{g}^{-1}$ ), and chicken feces (130 particles  $\text{g}^{-1}$ ). Macroplastics were also confirmed in the chickens' gizzards and crops. The authors suggested that the chickens' feeding and digestive processes further fragmented the ingested macroplastics. In addition, the presence of microplastics in soils has been observed to alter resident microbial respiration and other processes (Yang et al., 2018). Such "ecosystem" metabolic consequences require greater investigation in both terrestrial and aquatic environments.

Land-based microplastic sources are diverse and include landfills, wastewater solids and effluents, losses from industrial facilities (including plastics manufacturing), plastic agricultural mulch, polymer paints, and vehicle tire abrasion (Chae & An, 2018).

#### 3.1.1. Landfills and Dumps

Most plastics are disposed of in landfills in developed countries. These vary from secure, lined, and covered facilities to open trash piles that are later abandoned. However, even developed countries have had issues with off-site losses or mismanaged waste. For example, activities associated with the Fresh Kills landfill that served New York City were suggested as a major source of the medical waste (the so-called "syringe tide") that washed up on nearby beaches in 1987–1988 (Sheer & Moss, 2011). Historically, and in developing

countries today, many dumps have been located in low-lying areas, as such lands were deemed of limited value due to flooding (Brand et al., 2018). As sea level rises, these zones will be subject to greater flooding and erosion, contributing to additional release of plastic debris. Construction and demolition (C&D) landfills often serve as the repositories for debris following disasters such as hurricanes, tsunamis, and wildfires. These landfills are typically engineered and regulated less strictly than municipal landfills. C&D debris is presumed to be less hazardous than other wastes but may contain large amounts of plastics (e.g., furniture and insulation) with high additives levels. In the U.S. in 2002 alone, 610–780 million tons of C&D debris were generated, versus 214 million tons of municipal solid waste (Powell et al., 2015). Leachates and erosion from C&D landfills may thus also be a source of microplastics and additives to surrounding areas.

### 3.1.2. Burning of Plastics

Plastic trash and e-wastes are often burned under poorly controlled conditions liberating contaminants (Gullett et al., 2007; Asante et al., 2016). Releases of microplastics and additives as a result of these, as well as wildfires, have been inadequately evaluated. In the U.S. in 2016, 1.3 million fires occurred (<https://www.usfa.fema.gov/data/statistics/#causesR>). A total of 347 fires at waste and recycling facilities were reported in North America from April 2017 to 2018 alone (Fogelman, 2018). Additionally, wildfires are increasing in frequency worldwide with climate change (Keeley & Syphar, 2016). These fires engulf homes, businesses, and vehicles that contain abundant polymeric materials. The charred, disintegrating plastics may be transported offsite and into waterways. Airborne particulates are also produced and include plastic additives. For example, Ni et al. (2016) reported that airborne particulates and residual ash exhibited  $\text{mg kg}^{-1}$  concentrations of flame retardant polymer additives after plastic wastes were burned. Dust collected from New York City streets following the 2001 World Trade Center terrorist attack contained flame retardant polymer additives (Lioy et al., 2002). Resulting particulate matter may be small ( $<2.5 \mu\text{m}$ : PM<sub>2.5</sub>) and can penetrate deeply into respiratory tracts of air breathing organisms, including humans (Liu et al., 2016).

### 3.1.3. Tire Wear

Another source of microplastics to terrestrial ecosystems is from vehicle tire wear. Modern tires contain fillers (e.g., carbon black), additives, metallic and polymeric fibers, and natural and synthetic rubbers (primarily butadiene and styrene-butadiene polymers). Kole et al. (2017) estimated that each person in the U.S. generated  $4.7 \text{ kg year}^{-1}$  of tire wear microplastics, equivalent to 1.8 million metric tons  $\text{year}^{-1}$ . These authors hypothesized that tire wear may contribute 5–10% of global ocean plastics loading, as well as 3–7% of PM<sub>2.5</sub> in urban air. Transport of this material to waterways may occur via surface runoff, exacerbated by the impermeability of road surfaces. Alternatively, tire wear fragments enter sewer systems and then wastewater treatment plants. Locally, amounts released will vary depending on miles driven, climate, and topography. An estimate for the OSPAR catchment (essentially, the European countries bordering the North-East Atlantic Ocean) suggested that the amount of microplastics transported to local marine environments from tire wear was comparable to land-based litter. This was followed by that from paints, preproduction pellets, cosmetics, and laundry fibers. However, estimated ranges spanned orders of magnitude (OSPAR, 2017).

### 3.1.4. Paint and Coatings

Paints and surface coatings often contain polymers. Painted surfaces include structures, roadway markings, and vessels and are subject to weathering. Abrasive blasting (occasionally using microbeads) prior to repainting of surfaces will also generate microparticles. Paint often contain metal-based pigments (e.g., Cu and Zn). Takahashi et al. (2012) observed that up to 0.2% of the mass of cored sediments from the Plymouth estuary (UK) consisted of paint particles. Song et al. (2015) investigated microplastics in waters of Jinhae Bay, Korea. They reported that the abundance of paint particles exceeded those of other microplastic types and that size frequencies peaked in the 50 to 100  $\mu\text{m}$  range. The authors indicated that alkyd ship paint resins and poly (acrylate/styrene) from fiberglass resins were dominant polymers. They also noted that microplastics concentrated in the surface microlayer, a biologically important interface. Chae et al. (2015) published similar findings for the Incheon/Kyeonggi coastal region (Korea). Gove et al. (2019) observed that surface slicks contained a plastics-to-larval fish ratio that was sixtyfold higher than that of adjacent waters. They further estimated that such slicks represented only 8.3% of ocean surface habitat in a coastal Hawai'i ecosystem but contained 91.8% of floating plastic. Imhof et al. (2016) reported microplastics and paint particle loads in beach sediments from an Italian subalpine lake. They observed that the paint particles typically were smaller than other types of microplastics, mostly 1 to 50  $\mu\text{m}$ , likely due to their brittleness.



### 3.1.5. Microplastics From Clothes Washing

Habib et al. (1998) was among the first to suggest synthetic fibers, originating from textile washing, might be useful tracers of wastewater effluents and land-applied biosolids. Due to differences in media sampled and analytical methods, researchers do not yet agree on which microplastic types are most dominant in the environment. However, the International Union for the Conservation of Nature ranked releases from the laundry of synthetic textiles first, contributing 35% of the world ocean's microplastic burden (Boucher & Friot, 2017). Napper and Thompson (2016) estimated that over 700,000 fibers could be released from a 6 kg domestic wash load of acrylic fabric. Browne et al. (2011) sampled wastewater from washing machines and observed that a single garment could produce >1,900 fibers per wash load. Microplastics can then enter septic systems or be transferred to wastewater treatment facilities for additional treatment. However, in developing countries, textile wastewater may directly enter streams. Zambrano et al. (2019) observed lower biodegradation of synthetic fibers such as polyester in a simulated aquatic environment than naturally derived fibers, that is, cotton, polyester/cotton, or rayon. Biodegradation after 243 days of exposure was approximately 76% for cotton, 62% rayon, 40% polyester/cotton, but only 4% for polyester. Bajpai et al. (2011) indicated that microbial adherence to polyester fibers was low compared to cotton and polyester/cotton blend fibers.

Textiles can also trap airborne particulate pollutants. For example, Stapleton et al. (2005), detected flame retardant additives and triethyl phosphate (a polymer resin modifier used in unsaturated polyesters) in clothes dryer lint. Schreder and La Guardia (2014) detected several polymer additives not used in clothing (e.g., Penta-PBDE formulation used primarily in polyurethane foam) in laundry wastewater. They suggested that airborne microplastics adhering on clothing were transferred to laundry wash water. They also reported that hydrophilic polymer additives (e.g., chlorinated phosphate esters) in laundry wastewater exhibited <16% removal rates following wastewater treatment. In contrast, hydrophobic additives (e.g., PBDEs) had a >86% removal rate. The hydrophobic additives could eventually be introduced to soils via land-applied biosolids, while more water-soluble additives would enter receiving waters via effluent discharge. With respect to the latter, O'Brien et al. (2015) estimated that  $2.1 \text{ mg person}^{-1} \text{ day}^{-1}$  of organophosphate esters are transferred to treatment plants.

### 3.1.6. Wastewater Treatment

Industrial and domestic wastewaters contain microplastics and polymer additives derived from consumer products (Schreder & La Guardia, 2014). In affluent countries, wastewaters and storm water runoff from urban areas are typically routed to centralized treatment facilities. The treated effluents are eventually discharged to receiving waters. Some effluents, however, particularly in arid areas, are redirected for irrigation. Microplastics therein are then introduced to soils. Indeed, some wastewater treatment plants have instituted additional cleanup steps and have rebranded themselves "water reclamation facilities."

Microplastic fate during wastewater treatment is primarily influenced by particle densities. Most treatment schemes employ an initial screening of influent to eliminate macrodebris and settling to remove dense sand and grit. These byproducts are normally sent to a landfill. The next step (termed "primary") typically includes surface skimming and solids settling. Secondary treatment follows with aerobic digestion of labile organic matter and additional solids settling. A polymeric or inorganic flocculant is often added to improve particle sedimentation. Microplastics are sequestered into the settled solids to varying extents by these steps, with overall treatment removal rates of 90–99% in well-designed systems (Carr et al., 2016; Raju et al., 2018). Murphy et al. (2016) noted that most of the buoyant microplastics, including the majority of microbeads from personal care products, were entrained in the floating grease fraction. They observed that 78.3% of microplastics were removed during primary treatment, while secondary removed 20.1%. Exclusion of oil and grease and primary sludge from land-applied materials would thus reduce the amount of microplastics transferred to soils. Treatment may be less effective for microplastics <100  $\mu\text{m}$ . While facilities rarely implement steps aimed specifically at microplastics removal, interest is increasing. For example, Talvitie et al. (2017) evaluated several advanced options: discfilter, rapid sand filtration, dissolved air flotation, and membrane bioreactor.

Most microplastics that enter wastewater treatment works are sequestered in sewage sludge, which may be land applied to soils as a fertilizer. Land application economizes on landfill space and disposal costs. In the U.S., approximately 60% of wastewater sludge (also known as biosolids) is applied on agricultural fields, reclamation sites, or sold directly to consumers for use on gardens and landscaping. Mahon et al. (2017) reported microplastic burdens of 4,196 to 15,385 microparticles  $\text{kg}^{-1}$  in Irish sludges destined for land

application. Repeated applications on the same plot (common for fertilizers) increased proportionately the abundance of microplastics in receiving soils (Corradini et al., 2019). Nizzetto et al. (2016) estimated that the amount of microplastics added to European and North American soils via such applications exceeds the global burden now present in oceanic surface waters. Weithmann et al. (2018) observed that composts derived from household and commercial sources contained up to 895 microplastics  $\text{kg}^{-1}$ . The smallest microplastics examined by these authors were 1,000  $\mu\text{m}$ , so the actual abundances were likely much higher. Microplastics in soil may persist for decades or longer. Indeed, Zubris and Richards (2005) proposed their presence as a marker of past sludge applications. Plastic sheeting is widely used in agriculture to retain soil moisture and heat and block weed growth. Its breakdown is a source of microplastics to soils and later surface waters (Steinmetz et al., 2016).

Even when treatment facilities use techniques with high removal efficiencies or do not land-apply sludge, the massive water volumes of effluent discharged introduces substantial amounts of microplastics to receiving waters (Kay et al., 2018). Mason, Garneau, et al. (2016) surveyed 17 U.S. wastewater facilities (all practicing secondary treatment) and estimated that these were releasing on average >4 million microparticles per facility per day. They observed that tertiary filtration treatments (granular or biological aerated filter) were ineffective at further reducing microplastic discharges. Murphy et al. (2016) estimated that a secondary Scottish treatment plant (serving a population of 650,000) with a removal rate of >98% still released 65 million microplastics to receiving waters each day. Wastewater treatment plant discharges are often located on rivers, estuaries, and oceanic coastlines to take advantage of subsequent dilution and removal processes and thus are important sources of microplastics to these water bodies (Conley et al., 2019). McCormick et al. (2014) reported that microplastic concentrations (mean: 18 particles  $\text{m}^{-3}$ ) downstream of an urban wastewater treatment plant were comparable to maximum coastal concentrations after storm events.

Some treatment facilities employ biological aerated filters containing plastic “bio-beads” as high surface area substrates. The biofilm formed on these surfaces facilitate digestion of wastewater organic matter. These bio-beads may be similar in size to preproduction pellets (nurdles) used to manufacture plastic products. Turner et al. (2019) suggested that the release of bio-beads contributed to the plastic litter observed on western European beaches. They further hypothesized, based on elemental analyses (e.g., Br, Sb, and Pb), that some of the bio-beads might be derived from recycled e-waste plastics containing toxic additives such as flame retardants. Wastewater treatment facilities may also contribute to the growth of novel bacterial biofilm assemblages on microplastics discharged. For example, McCormick et al. (2014) observed significant colonization of microplastics by wastewater-associated organisms, including some plastic decomposing (e.g., *Pseudomonas*) and potentially pathogenic taxa such as *Campylobacteraceae*. Microplastics colonized with such microbes may introduce them to receiving waters if they escape wastewater treatment facilities.

It is important to note that most published studies focus on microplastic fate in advanced treatment facilities. Worldwide, many locales have no treatment or primary only. In addition, large rainfall events may result in wastewaters bypassing treatment altogether, especially at facilities with combined sewer overflows (see discussion on storms). Even in affluent countries such as the U.S., waivers allowing primary-only treatment have been issued to facilities discharging into open or fast-moving marine waters, for example, in California, Hawaii, Guam, Virgin Islands, Alaska, and Puerto Rico. Cities in several Canadian provinces also discharge to the ocean following only crude treatment (e.g., Johannessen et al., 2015). As another example, Antarctica is largely pristine. Yet prior to 2003, domestic wastewater was simply macerated and then discharged directly to adjacent McMurdo Sound, under the premise that constituents therein were biodegradable and subsequently well diluted. Even after implementation of advanced wastewater treatment in 2003, polymer additives in indoor dust and treated effluents from this facility, as well as their presence in sediments and biota of the neighboring environment, were detectable (Hale et al., 2008). Microplastics likely served as a carrier for at least some of these.

### 3.2. Direct Releases to Surface Waters

Plastics may enter surface waters directly via fishing and aquaculture, intentional disposal from vessels, and storm-related debris. Nets, lines, floats, and traps may contain plastics and synthetic fibers. Although these are designed to resist weathering, over time they will degrade. Lost fishing gear retains its capacity to ensnare (“ghost fish”) biota for years. Wilcox et al. (2015) postulated that >6 million tons are lost annually. Lebreton et al. (2018) estimated 45,000 to 129,000 tons of plastic were floating within the Great Pacific Garbage Patch.

About 46% of that consisted of lost fishing nets and 8% of microplastics. Fragments of derelict nets and lines can also catch on obstructions such as reefs, further damaging marine life. These continuously degrade, forming secondary microplastics. While many wild-caught fisheries are in decline, aquaculture has expanded at an annual rate of 5.8% over the last 30 years (Oyinlola et al., 2018). Aquaculture is typically done near shore and is generally better monitored than wild-caught fisheries (Schoof & DeNike, 2017), yet some associated gear will be lost and deteriorate.

Plastics also enter waters via cargo lost overboard and materials discarded or otherwise lost on or adjacent to shorelines. The International Convention for the Prevention of Pollution from Ships (MARPOL) Annex V entered into force in 1988 to reduce the discharge of garbage (including plastics) to the ocean (<http://www.imo.org/en/OurWork/environment/pollutionprevention/garbage/Pages/Default.aspx>). However, it is voluntary and only applies to larger vessels on international voyages. The 2005 MARPOL Annex IV restricts the discharge of sewage from ships. As noted previously, wastewaters contain microplastics. The International Maritime Organization states: "It is generally considered that on the high seas, the oceans are capable of assimilating and dealing with raw sewage through natural bacterial action." Dumping of sewage is allowed when the ship is in operation and has an approved sewage treatment plant or when the ship is discharging comminuted and disinfected sewage using an approved system more than three nautical miles from the nearest land. It allows untreated sewage to be discharged if the ship is more than 12 nautical miles from the nearest land.

In some cases, plastics and construction debris have intentionally been deployed in water bodies, for example, for the creation of artificial reefs to attract finfish and shellfish (Collins et al., 1995). From 1970 to 1990, about 1 million waste automobile tires were sunk in coastal zones (Faverney et al., 2010). In some cases, tires were chipped to increase surface areas, placed in mesh bags, and then anchored down. Unfortunately, some of these materials broke free, damaged nearby reefs, and accumulated on beaches (Sherman & Spieler, 2006). Some were later removed at substantial cost; in other cases the materials were abandoned. After loss, microplastics were surely created as the materials were abraded and weathered.

Preproduction pellets (i.e., nurdles) and other microplastics have been observed on shorelines near plastics manufacturing facilities or due to riverine, lacustrine, estuarine, and marine circulation patterns (Antunes et al., 2013; Browne et al., 2010; Klein et al., 2015; Zbyszewski & Corcoran, 2011). In the northeast Atlantic preproduction pellets have been suggested as the fourth largest direct source of microplastics to surface waters, behind tire wear, land-based litter, and deteriorating paints (OSPAR, 2017). Pellets and resins may be lost from manufacturing facilities and during shipping activities. Loss rate estimates vary from about 0.01 to 0.1% of production and shipping (OSPAR, 2017). A program known as Operation Clean Sweep® has been implemented in North America and the UK by the plastics supply chain to reduce losses of such materials ([http://www.opcleansweep.eu/wp-content/uploads/2019/03/OCS\\_A4\\_Report\\_2018\\_ONLINE.pdf](http://www.opcleansweep.eu/wp-content/uploads/2019/03/OCS_A4_Report_2018_ONLINE.pdf)).

Episodic events such as storms scour shorelines and periodically inundate urbanized areas. Floodwaters carry deposited debris into receiving waters. Storm waters also can inundate treatment plants resulting in untreated wastewater entering receiving waters (Kiaghadi & Rifai, 2019). Nonetheless, few studies have examined the role of extreme weather on the fate of plastics. J. Wang, Lu, et al. (2019) reported that microplastic abundances increased 40% in Sanggou Bay, China, following a typhoon. Hurley, Woodward, et al. (2018) detailed the existence of several riverine sediment microplastic hotspots, with concentrations as high as 517,000 particles  $\text{m}^{-2}$ . After a series of floods, they concluded that about 70% of the existing in-place microplastics (equivalent to about  $0.85 \pm 0.27$  tons or  $43 \pm 14$  billion particles) were exported downstream towards the Irish Sea.

Devastating tsunamis can travel kilometers inland, and receding waters introduce massive amounts of debris into oceans. For example, the 2011 Great Japan Tsunami injected an estimated 5 million tons of debris into the Pacific (Murray et al., 2018), approximating the estimated amount of plastics entering the oceans each year worldwide. Lebreton and Borrero (2013) calculated that this single event was >3,000 times the average yearly amount of land-based litter contributed by all of Japan and exceeded by thirteenfold the amount of plastic currently in Atlantic surface waters. Once in the ocean, debris was fractionated by physical processes. Dense debris sunk near shore. Based on modeling and observations, most of the smaller floating debris eventually entered the Pacific "garbage patch" (Maximenko et al., 2018). Substantial debris reached North America and Pacific island shores. During peak deposition periods, Murray et al. (2018) estimated that

debris strandings increased tenfold along the North American west coast. Some large windage debris was steered farther north. Much will never be recovered and will fragment in situ, increasing microplastic loads.

### 3.3. Atmospheric Transport

Little research has examined atmospheric transport of microplastics to date. As the oceans cover over 70% of the globe, atmospheric deposition of microplastics to marine environments is likely substantial. Recently, Dris et al. (2016) calculated atmospheric deposition of synthetic fibers, amounting to 3 to 10 metric tons each year, on a 2,500 km<sup>2</sup> area encompassing the footprint of greater Paris, France. Deposition reached 355 particles m<sup>-2</sup> day<sup>-1</sup>. Small fibers, 7 to 15 μm in diameter, were the most abundant. While rain events increased deposition, there was not a direct correlation with rainfall amount. Microplastic deposition was greater at urban than suburban sites. K. Liu, Wang, et al. (2019) reported slightly higher airborne microplastics levels for Shanghai. They observed that fibers constituted 67% of microplastics observed and estimated that Shanghai residents inhaled about 21 microplastic particles day<sup>-1</sup>. Allen et al. (2019) examined atmospheric deposition in a remote Pyrenees (France) alpine catchment and found rates (as high as 365 microplastics m<sup>-2</sup> day<sup>-1</sup>) comparable to the above urban areas. They suggested that some microplastics may be transported thousands of kilometers and across oceans and that rain and snowfall events facilitated deposition. Most fragment particles were <50 μm, but fibers were predominantly 100 to 300 μm in length. Unlike other studies, they reported that polystyrene was a dominant polymer type in deposition samples.

## 4. Microplastics in Freshwaters

After release to the terrestrial environment, plastics may be transferred to wetlands, lakes, and rivers. Human population densities are higher near water bodies, as they provide valuable transportation routes and suitable water for irrigation, industrial application, and consumption. The world's rivers are especially vulnerable to pollution, altered hydrology, and introduction of invasive species (Best, 2019). The occurrence of microplastics in freshwaters has been reviewed (Eerkes-Medrano et al., 2015; Horton et al., 2017; Li et al., 2018; Wagner et al., 2014). Surface runoff and atmospheric deposition transfer plastic debris and microplastics within the drainage area into freshwater receiving systems. Plastic burdens in these aquatic systems, along with direct dumping and littering, eventually move downstream and enter estuaries and coastal seas. The importance of rivers as conduits of plastic waste was suggested by Lebreton and Andrady (2019). They estimated that 91% of mismanaged plastic wastes is transported via watersheds larger than 100 km<sup>2</sup>. They further suggested that >25% of global wastes have been discarded into 14 large (>1,000,000 km<sup>2</sup>) riverine watersheds in North America (Mississippi, Nelson, and Saint Lawrence), South America (Amazon and Paraná), Africa (Congo, Niger, Nile, and Zambezi), Europe (Volga and Lena), and Asia (Amur, Yangtze, and Ganges).

### 4.1. Microplastics in Lakes and Rivers

Globally, no obvious regional patterns in microplastic concentrations among lakes have yet been established, likely because burdens are impacted from local sources and sampling coverage is sparse. For example, no published data on microplastics in the largest freshwater lake in the world, Lake Baikal (Russia), was found. However, Battulga et al. (2019) presented results on plastic debris along the shore of the Selenga River, a Baikal tributary, in Mongolia. Polystyrene foam, believed to be from local sources, was observed to be the major type of debris. The Laurentian Great Lakes contain 21% of the world's surface fresh water (slightly less than Baikal) and serve as a major drinking water source for U.S. and Canadian residents. Eriksen et al. (2013) estimated that average abundance of microplastics in Lake Superior, Huron, and Erie was 43,000 particles km<sup>-2</sup> (equivalent to 0.27 particles m<sup>-3</sup>). However, Baldwin et al. (2016) observed an order of magnitude higher level (466,000 particles km<sup>-2</sup>, equivalent to 2.9 particles m<sup>-3</sup>) near several large cities. Eriksen et al. (2013) also reported that Lake Erie, which has a greater number of major cities with populations exceeding 100,000 in its immediate watershed than the other Great Lakes, was more polluted than Lake Superior or Huron. They also noted that microplastics <1 mm accounted for 81% of total particles in the Great Lakes. Mason, Kammin, et al. (2016) reported that surface waters of Lake Michigan had an average abundance of 17,000 particles km<sup>-2</sup>. Ballent et al. (2016) evaluated microplastics in sediments of Lake Ontario. They found an average of 760 particles kg<sup>-1</sup>. Higher levels were apparent in sediments near urban centers such as Toronto, Canada. Etobicoke Creek (a tributary of Lake Ontario and home to several plastics manufacturers and distributors) contained the highest concentration: 28,000 particles kg<sup>-1</sup>. In Lake



Winnipeg surface waters, higher abundances of microplastics occurred in nearshore waters than offshore waters, demonstrating the influence of land-based sources (Anderson et al., 2017). This lake is located in a relatively undeveloped area of Manitoba, Canada, and is the eleventh largest freshwater lake in the world.

Microplastics have also been reported in European lakes, wherein concentrations were 2.68 to 3.36 and 0.82 to 4.42 particles  $\text{m}^{-3}$  in surface waters of Lake Chiusi and Lake Bolsena (Italy), respectively (Fischer et al., 2016). The authors noted that high winds affected the surface water distribution of microplastic fragments, but not fibers, demonstrating another variable that should be considered when sampling. An average microplastic concentration of 0.5 particles  $\text{m}^{-3}$  was reported in Swiss lakes, with individual concentrations ranging from 0.06 particles  $\text{m}^{-3}$  in Lake Zurich to 1.2 particles  $\text{m}^{-3}$  in Lake Maggiore and Grand Lac Geneva (Faure et al., 2012). In Asia, microplastics were detected in remote Lake Hovsgol, in Mongolia (Free et al., 2014). In Lake Taihu (China) the concentrations of microplastics varied by location, with lower concentrations in the central part (0.3 particles  $\text{m}^{-3}$ ) and higher levels in the northwestern (1.1 particles  $\text{m}^{-3}$ ) and southeastern (2.5 particles  $\text{m}^{-3}$ ) sections (Su et al., 2016).

Lakes generally have large surface area to shoreline ratios, resulting in dilution. For example, microplastic abundances decreased with increasing lake surface area for the Great Lakes mentioned above, with individual mean concentrations of 0.66, 0.034, and 0.02 particles  $\text{m}^{-3}$  for Lake Erie (surface area: 26,000  $\text{km}^2$ ), Huron (60,000  $\text{km}^2$ ), and Superior (82,000  $\text{km}^2$ ), respectively (Eriksen et al., 2013). This pattern also follows the population densities and degree of industrialization in the associated watersheds. As further evidence of the dilution capacity occurring in lakes, Baldwin et al. (2016) determined that microplastic concentrations in the surface waters of tributaries exceeded those of the Great Lakes; that is, the mean concentration was 4.2 particles  $\text{m}^{-3}$  in 29 tributaries, while only 0.27 particles  $\text{m}^{-3}$  in the Great Lakes. This underlines the role of the tributaries as sources or conveyors of microplastics. About 72% of the plastic particles in the tributaries were <1 mm, comparable to 81% in the Great Lakes. Microplastic concentrations were positively correlated with the levels of urbanization and runoff in the watersheds (Baldwin et al., 2016), supporting the influence of both anthropogenic activities and hydrological conditions on the occurrence of microplastics in both the tributaries and lakes themselves.

As land-based sources of plastic debris dominate, rivers act as major receptors and conduits of debris to lacustrine systems (Schmidt et al., 2017). Disparities between microplastic concentrations at different sampling sites may be due to point sources or the sampling methods employed. Dris et al. (2015) observed high microplastic concentrations in rivers (3–106 particles  $\text{m}^{-3}$ ), attributed to urban inputs such as treatment plant discharges. Kapp and Yeatman (2018) reported microplastic concentrations in surface waters from the 1735 km Snake and 523 km lower Columbia Rivers (U.S.). They reported higher levels in areas adjacent to agricultural areas, possibly due to the use of biosolid fertilizers or plastic mulches. Recreational areas also exhibited elevated levels. Somewhat surprisingly, a correlation of microplastic levels with human population densities was not observed. Microplastic concentrations were negatively correlated with water velocity. The authors hypothesized that dams might contribute to accumulations of less dense microplastics, as did K. Zhang et al. (2017) investigating distributions of microplastics in the Yangtze River near the Three Gorges Dam in China. McCormick et al. (2014) observed microplastic concentrations increased from upstream ( $1.94 \pm 0.81$  particles  $\text{m}^{-3}$ ) to downstream ( $18 \pm 11$  particles  $\text{m}^{-3}$ ) sites in the Illinois River, a tributary of the Mississippi. Microplastic concentrations here varied as the river flowed through multiple cities (McCormick et al., 2014). Another example is the Rhone River (Switzerland), where a higher mean concentration was found near Geneva (0.29 particles  $\text{m}^{-3}$ ) than Chancy (0.13 particles  $\text{m}^{-3}$ ; Faure et al., 2012). Plastic waste in river waters may also strand on shorelines, embed in sediment, be ingested by organisms, or be carried downstream. However, floods may later resuspend microplastics in sediments and recapture shoreline debris.

#### 4.2. Riverine Input of Microplastics to Coastal Zones, Estuaries, and Oceans

Although the transition between the sources of plastic pollution (inland streams and rivers) and their sinks in coastal zones and the oceans is critically important, less research has investigated this transition area than the remote ocean gyres. Several models have been developed to estimate the export of debris from rivers to coastal zones and the oceans. Lebreton et al. (2017) estimated that 1.15 to 2.41 million tons of plastics are released to the oceans through river-fed estuaries each year. These authors also considered the impact of dams and related water retaining structures on transport of microplastics. Schmidt et al. (2017) reported a similar range of annual plastic inputs: 0.47 to 2.75 million tons. The concept of mismanaged plastic waste

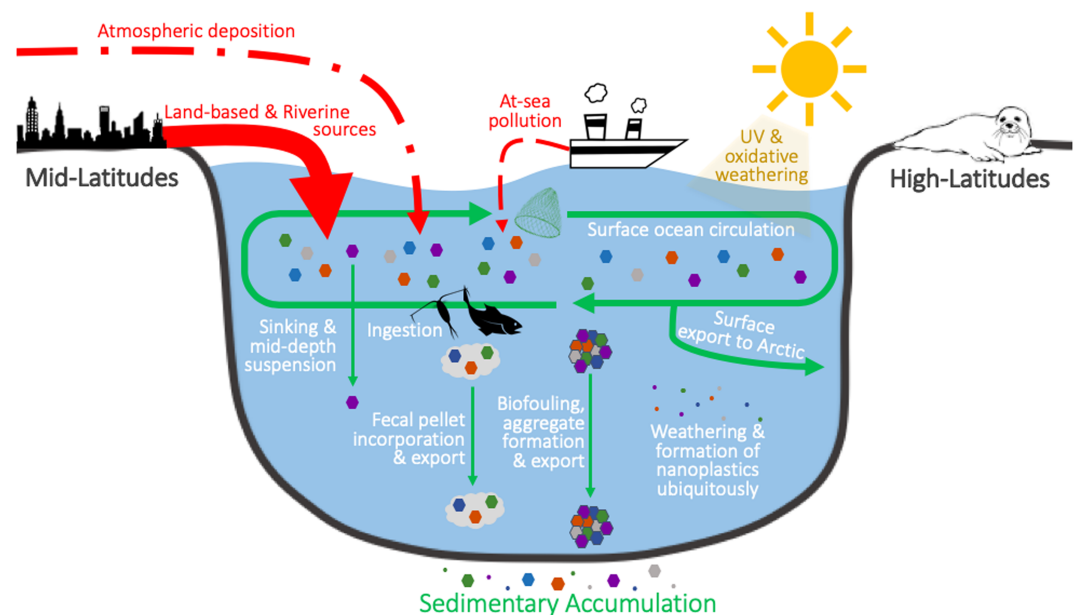
(Jambeck et al., 2015) has been used to estimate riverine plastic inputs (Lebreton et al., 2017), although additional, updated metrics are needed to improve models.

It is likely that coastal water bodies (e.g., bays and estuaries) are even more polluted than rivers due to multiple input sources and circulation patterns in these semienclosed basins. For example, the Beiji River of South China was reported to contain a mean concentration of  $0.56 \pm 0.45$  particles  $\text{m}^{-3}$  compared to 0.11–68 particles  $\text{m}^{-3}$  in Xiangxi Bay (Tan et al., 2019). Zhao et al. (2014) observed surface water microplastic levels as high as 4,137 particles  $\text{m}^{-3}$  in the Yangtze Estuary (China). Dominant forms observed were fibers, but unfortunately, the authors did not report polymer composition. Cheung et al. (2016) observed much higher microplastic concentrations during the wet than the dry season in Hong Kong waters near the Pearl River Estuary. These authors suggested that if microplastic concentrations exhibit seasonal variability, seasonality must be considered to reduce bias in estimating overall riverine inputs of microplastics. Yonkos et al. (2014) reported positive correlations between the plastic concentrations in four estuarine rivers of the Chesapeake Bay and both population density and extent of urbanization within the watersheds. A 1-year survey of the Nakdong River (the longest river in South Korea) with four sampling time points found riverine inputs of microplastics of 53 to 118 tons  $\text{year}^{-1}$ , with highest microplastic concentrations in the wet season (Eo et al., 2019). Unfortunately, there are few studies to compare results with, as most sampling campaigns have been conducted over only a few months with fewer than four consecutive field measurements. As such, current modeling exercises have largely relied on fragmented data to estimate riverine plastic inputs and incorporate insufficient validation (Schmidt et al., 2017).

While the presence of microplastics in estuaries has been documented (Gray et al., 2018; Pazos et al., 2018; Yonkos et al., 2014; Zhao et al., 2015), our understanding of their distributions and processes remains limited. However, it is likely that principles gleaned from studies of naturally occurring particulates of comparable densities are applicable, especially as microplastics weather and accumulate biofilms. In terms of behavior after release, Hoellein et al. (2019) observed that microplastics mimicked natural particles in terms of deposition in an experimental stream. They argued that existing studies on particulate organic matter could provide insights on microplastic behavior and fate. They observed that high-density microplastics and those with “sticky” biofilms settled the quickest, while fibers remained suspended longer and exhibited less biofilm formation. The most prevalent types of polymer types reported in estuaries have been polyethylene, polypropylene, and polystyrene (Sadri & Thompson, 2014). In part, this is because most studies have focused on shorelines and surface sampling, which preferentially collect these low-density polymers. In a South American study, Acha et al. (2003) suggested that a benthic estuarine salinity front acted as a seaward barrier to and concentrated riverine-derived macroplastic debris. They also reported that this zone was a focal point for finfish and shellfish, as well as zooplankton. Cohen et al. (2019) sampled and modeled the surface distributions of buoyant microplastics in the Delaware Bay (USA) estuary and observed substantial spatiotemporal variability. They noted highest concentrations near the estuarine turbidity maximum. Browne et al. (2010) evaluated the roles of wind, waves, and tides on the fate and deposition of shoreline plastic debris in a UK estuary. They observed greater amounts of plastics at downwind sites and higher amounts of denser microplastics in sediments.

Both physical and chemical interactions may affect microplastic fate in waterbodies (including bays and estuaries) where flora and fauna, as well as salinity and pH gradients, exist. In the coastal zone, Li et al. (2018) observed that polystyrene microplastics, associated with local mollusk aquaculture, were dominant pollutants in Quinzhou Bay (China) and that the dense vegetation in the system trapped plastic debris. Shellfish are also efficient at removing suspended particulates and are common inhabitants of such environments. Therefore, filtration and subsequent deposition to sediments via feces or pseudofeces may be important microplastic removal mechanisms (Zhao et al., 2018). Microplastics may exhibit surface charge alterations, especially after weathering and biofouling (Paul-Pont et al., 2018). Such outcomes may result in enhanced aggregation and flocculation, with resultant sinking and ultimately deposition in sediment. Interactions between charged plastic surfaces may also occur with metals and organic chemical species (Holmes et al., 2014). Chemical constituents of microplastics may also be transported from terrestrial runoff to oceanic systems via estuaries. For example, Amamiya et al. (2019) reported styrene oligomers, derived from polystyrene, entered Tokyo Bay from land-based sources as a function of monthly precipitation.

Overall, the transport of plastic debris from land to oceans is a complex process and affected by a variety of factors, including human population density, urbanization, per capita income, hydrological conditions,



**Figure 5.** Global ocean distribution of microplastics (portrayed simplistically as multicolor hexagons). Inputs of microplastics are shown in red, while processes that control their distribution and export from the surface are in green. Photodegradation (the most rapid weathering process) of microplastics will be exacerbated in exposed terrestrial situations and the surface ocean. Ultimately, microplastics will accumulate in deep ocean sediments.

waste management infrastructure, and living standards. More integrated models are desirable, to better constrain global riverine inputs and reflect the state of plastic pollution. This should be supported by long-term monitoring programs, better sampling strategies to encompass variability (e.g., tidal oscillations), and improved model calibration and validation.

## 5. Fate of Microplastics in the Ocean

Media reports of the “Great Pacific Garbage Patch,” notorious as an area of floating trash “twice the size of Texas” in the Pacific Ocean sparked global interest in plastic pollution. Trawling the water’s surface with 333  $\mu\text{m}$  mesh nets, Moore et al. (2001) reported the mass of microplastic in samples from the North Pacific gyre was six times that of coincident plankton. Although this galvanized global discussion of marine debris, reports existed as early as 1965 (Ostle et al., 2019). Due to their size, bathymetry, and position in the hydrological cycle, oceans serve as a sink for plastic debris and other persistent pollutants. Basic physical oceanographic considerations (Ekman transport, geostrophy, etc.) explain why plastic debris accumulates within the more quiescent zones of oceanic gyres, while more complex processes determine the distribution of marine plastics in less obvious locales, including deep sea sediments and ice sheets (Figure 5). Research combining modeling and observational data has been employed to better understand the distribution and trends of microplastics in oceans, bays, and estuaries. As an example of this approach and to illustrate the potential gravity of the situation, Isobe et al. (2019) predicted that the weight of pelagic microplastics around the Pacific Ocean subtropical convergence zone would double by 2030 and quadruple by 2060. They postulated that the latter might pose health risks to resident marine organisms based on available, albeit limited, toxicological studies. The authors incorporated major assumptions concerning microplastic particle size, behavior, and their environmental persistence into their model. Clearly, research to refine such assumptions is crucial.

### 5.1. Theoretical and Empirical Models of Microplastics in Surface Oceanic Waters

Due to the vastness of the world’s oceans, theoretical and empirical models have been developed to predict where microplastics will accumulate. Early models used to predict marine debris distribution were designed to track ocean currents. However, these were modified by Maximenko and Niiler to explain hot spots of plastic accumulation (“Ocean Debris”, 2008). Their model identified the five ocean gyres (North Pacific, South Pacific, North Atlantic, South Atlantic, and Indian) as major destinations of marine debris.

Since then, more elaborate models of surface microplastic distribution have been developed. Lebreton et al. (2012) used predictions of plastic input into the oceans, combined with Lagrangian particle tracking and surface ocean circulation models to estimate microplastic distribution. They forecasted that debris will accumulate in the five gyres, with higher concentrations in the Northern Hemisphere (e.g., North Pacific Subtropical Convergence Zone). They also highlighted the importance of marine debris source and showed that debris may accumulate closer to shore (instead of only in the gyre patches), depending on the point of origin. The conclusions drawn by these authors corroborate another theoretical model, focused on North Pacific Ocean circulation patterns, and suggested that convergence patterns could result in amplified marine debris accumulations in certain areas (Howell et al., 2012). Another Lagrangian drift model was used to understand the movement of plastic debris in the North Sea. Using wind and current data, Neumann et al. (2014) projected the fate of microplastics from a hypothetical release of plastic pellets after nine years. The authors noted that seasonal differences factored into the model accurately predicted the different concentrations of pellets along the coastline over time. It further suggested that the coastal ocean patterns may be influenced in the short term (seasonally), while the open ocean debris would be affected over longer time scales. Unfortunately, the authors found that hindcasting the model did not accurately predict plastic source(s).

An important aspect of theoretical models for forecasting marine debris circulation pertains to the connectivity of surface oceans, or the balance between debris attraction to or dispersion from a central area, such as a gyre. Van Sebille et al. (2012) were among the first to use observational data to model the spread of plastic debris in the ocean. These authors combined data from the Global Drifter Program and applied a particle-trajectory tracker approach to predict where the particle would travel over seasonal, yearly, or multiyearly time frames. They confirmed the existence of five major ocean garbage patches but also predicted another in the Barents Sea. In addition, they used the model to track the “leakiness” of the garbage patches. They found that the patches are likely to be more dispersive than would have been projected from linear ocean circulation/Ekman theory, highlighting the importance of combining observational data with models. Froyland et al. (2014) used a Markov chain model to observe where in space surface water (and microplastics) may be sequestered over time. The authors highlighted the importance of understanding surface ocean circulation as a three-dimensional system instead of a two-dimensional plane and included ground truthed upwelling and downwelling observations in their model. They found that some small, uniquely shaped areas are likely to attract and keep ocean pollution “forever.” Contrary to intuition, these attracting regions are in the general area of ocean gyres but do not directly overlap with them, extending westward in the Southern Hemisphere and southward in the North Atlantic. Other regions that theoretically only lose debris over extended periods are found in the remainder of the five primary ocean gyre regions and in some coastal zones. The authors point out that this model may be valuable in understanding the long-term fate of marine debris, which cannot readily be accomplished on an observational time scale.

A number of researchers have used field-collected microplastics data to create empirical models. Goldstein et al. (2013) surveyed microplastics in the North Pacific Subtropical Gyre. They indicated the plastic densities correlated with the physical parameters of the water body. Empirical modelers have also utilized satellite observations. Sherman and Van Sebille (2016) combined empirical data on floating marine debris from satellite observations and sea surface trawls to estimate current plastic locations and project plastic densities in those locations until 2025. A goal of this study was to predict optimal removal locations for sea surface microplastics. Contrary to data highlighting plastic concentrations in the North Pacific Subtropical Gyre, the modeled optimal removal location was closer to the east coast of China, in part due to the high debris output from this region.

Another study tracked the movement of drogued and undrogued drifters as a proxy for marine debris (Beron-Vera et al., 2016). The authors found that undrogued drifters naturally moved toward areas where marine debris accumulates but point out that this could not be explained by Ekman surface transport alone. Rather, the drifter location was motivated by inertial properties of the drifter (controlled by finite size and buoyancy). They elucidated this through known properties of drifters but proposed that this could be applicable to all marine flotsam. In fact, recent work highlights that debris models often depend too much upon Ekman transport and geostrophic current patterns and fail to include processes such as wave-induced Stokes drift in their estimations (Biaosoch et al., 2019; Onink et al., 2019). By comparing models that include or omit wave-induced Stokes drift, Onink et al. (2019) found that existing models depending upon Ekman



and geostrophy are appropriate for most ocean basins, except for the Arctic. These authors also traced the transfer of microplastics between ocean basins, showing that those that originate in the North Pacific likely will remain there, while plastics originating in other basins are more likely to disperse to other basins. The exchange reported between the North Atlantic and Arctic oceans, models that include wave-induced Stokes, and thermohaline circulation patterns support the Arctic as a destination for surface microplastic pollution (Cózar et al., 2017), discussed later as a sink for ocean microplastics.

The majority of microplastic distribution models have focused on open ocean and gyre dynamics rather than the coastal zone. Zhang et al. (2017) combined the physical characteristics of microplastics and factors affecting coastal ocean dynamics (e.g., wind, waves, and tides) to develop a model for microplastics near coastlines. Models of debris distribution were also completed for the Mediterranean Sea, using Lagrangian drift models (Mansui et al., 2015). These data identified three zones that may be short-term hot spots for debris accumulation and additional areas that were unlikely to see debris accumulation on most time scales. In the long-term, however, the dynamical processes of a semienclosed ocean basin made it so that there was no area(s) projected to maintain debris on the oceanic time scales identified in Froyland et al. (2014). This could be attributable to assumptions of the model, however, including the unrealistically dense and homogeneous starting distribution of microplastics, as detailed by the authors.

Due to the complexity of modeling coastal ocean and enclosed basin dynamics, microplastic tracking in the coastal zone has been largely dependent upon observational studies. In a study of the northwestern Mediterranean Sea, Collignon et al. (2012) observed that 90% of the stations surveyed contained microplastics. The concentration of microplastics was lower following strong wind events, suggesting that wind plays a role in vertical mixing of microplastics out of surface waters. In a surface water study of microplastics in the North Yellow Sea, China, microplastics were observed at all 19 study sites, with an average concentration of  $545 \text{ items m}^{-3}$ , which is about average for other surface water studies in the region (Zhu et al., 2018). These authors attributed the higher abundance of these microplastics in the northern than the southern portion of the bay to prevailing wind patterns and proximity of developed land. Overall, these and other studies suggest that microplastics in coastal surface waters are at least as ubiquitous as in open ocean waters, yet modeling their distribution is complicated by local source hot spots, wind and precipitation patterns, morphology, and complex water circulation.

In a recent meta-analysis of microplastic polymer distribution, Erni-Cassola et al. (2019) confirmed that most polymers observed in surface waters were polyethylene and polypropylene, low-density polymers common in single-use products. This is true of coastal zones as well; for example, polyethylene dominated surface water in the aforementioned study of the North Yellow Sea (Zhu et al., 2018). Deeper in the water column, denser particles (e.g., polyester, polyamide, and acrylics) may dominate (Erni-Cassola et al., 2019). Recently, sampling protocols have begun to use smaller sampling mesh sizes, whole water samples, or more sensitive detection schemes, allowing determination of smaller microplastics. These changes have revealed that fibers are often more abundant than fragments in some aquatic systems (Barrows et al., 2018; Carr, 2017). Such fibers generally derive from textiles, rope, or netting and can include polyethylene, polyamide (nylon), and polyester. Polymer categorization is useful as it provides insight into the potential source of the plastic and explains distribution based on density. Yet other complexities, such as additive composition or biofilm presence, may have substantial impacts on microplastic fate, and effects are often unexplored.

## 5.2. Export of Microplastics From the Surface Ocean to Deep Waters and Benthos

Some models had predicted that there should be up to 100 times more plastic in the surface ocean than has been commonly reported (Cózar et al., 2014; Geyer et al., 2017; Jambeck et al., 2015). Early hypotheses for the location of this “missing plastic” included degradation into minute fragments below size detection limits, biofouling (which may lead to sinking), stranding of debris, ingestion by marine species, and incomplete observations (Cózar et al., 2014). Now, this “missing plastic” is primarily thought to be in the nanoplastic or microplastic size range and has motivated research on their fate beyond surface waters.

The Arctic Ocean (specifically, the seas east of Greenland), a site of deep water formation in thermohaline circulation models, and has been proposed as a “dead-end” for microplastics. Based on ocean circulation and Lagrangian drift models, Cózar et al. (2017) speculated that the Greenland and Barents Seas (sites of

North Atlantic Deep Water formation) likely accumulate debris. Field sampling by these authors confirmed that microplastics were abundant in these two seas compared to other portions of the Arctic Ocean. The type, size, and morphology of the plastics suggested that they were highly weathered and had traveled long distances on the surface ocean prior to reaching the Arctic. Researchers have hypothesized that once reaching the Arctic, microplastics may be vertically exported to the benthos. Kanhai et al. (2019) collected and evaluated sediment cores from the Arctic Central Basin for the presence of microplastics. They observed that 7 of the 11 samples contained microplastics (all  $<100\text{ }\mu\text{m}$ ), with a concentration of 0–200 particles  $\text{kg}^{-1}$ . These results may not be impressive at first glance, but considering the experimental limitations (including sample size and volume and extraction protocol), this study showed that microplastics are likely widely abundant in Arctic sediments. Bergmann and Klages (2012) previously observed a more than doubling of plastic and other anthropogenically derived macrodebris on the Arctic seafloor between 2002 and 2011 at the HAUSGARTEN observatory ( $79^{\circ}\text{N}$ ). They postulated that the increase might be related to receding sea ice.

Microplastics have been observed in deep-sea sediments from a variety of other locations, including the North and South Atlantic, Southern Ocean, Mediterranean Sea, and Indian Ocean. Although they did not determine polymer type, Van Cauwenberghe et al. (2013) found microplastics in 5 of 11 sediment samples, originating from 1,176 to 4,843 m in depth and between 44 and  $161\text{ }\mu\text{m}$  in size. In another study, Woodall et al. (2014) recovered microplastics from all 12 sediment samples collected in deep sea locations. All were fibers and mostly polyester or acrylic polymers, as would be expected based on their densities (Erni-Cassola et al., 2019). Fibers of cellulose-derived rayon were also detected. Concentrations ranged from 1.4 to 40 pieces per 50 ml of sediment. These authors also tested four deep-sea corals and found microfibers in all samples using a binocular microscope and entomological pin. It is not surprising that organic matter is reaching these deep-sea plains and trenches, as it has been found at surprising concentrations in other abyssal locations, such as the Atacama trench (Danovaro et al., 2003). These results contradict preconceived notions of the buoyancy of plastics and their propensity to sink to great depths.

Mechanisms have been proposed to explain the export of microplastics from surface waters to sediments, including increased density from biofouling. In a controlled study, Fazey and Ryan (2016) allowed biofilms to form on plastics of different sizes (5–50 mm) and observed sinking rates. They found that the smaller plastics sank faster than larger particles, yet all sank within the 12-week study period. Biofilm formation, however, is a highly variable process. As such, these results may not apply to all systems, particularly oligotrophic ocean gyres. Kooi et al. (2017) developed a model of microplastic vertical transport based on biofouling and water conditions. Their model suggested that smaller biofilmed microplastics ( $<1\text{ }\mu\text{m}$ ) will settle faster than larger ones and are less likely to resurface. Contrarily, particles from 1 to  $5\text{ }\mu\text{m}$  may oscillate in the middle water column, suggesting that some microplastics may be “lost” to the middepths, and neither retained in the surface water or sinking to greater depths. This correlates with previously discussed observational evidence that microplastics found in the deep sea were smaller than  $200\text{ }\mu\text{m}$  (Van Cauwenberghe et al., 2013; Woodall et al., 2014). Yet, the model also specified that particles  $<10\text{ }\mu\text{m}$  were likely to exit surface waters and remain suspended in the water column because of their extremely slow sinking rates (Kooi et al., 2017). This is a theoretical prediction yet useful for understanding the dynamics of microplastic sinking following biofilm growth. Choy et al. (2019) sampled microplastics between 100 and  $5,000\text{ }\mu\text{m}$  in water along vertical transects off Monterey Bay, California, using remotely operated vehicles. They observed maximum concentrations between 200 and 600 m. The microplastics observed were dominated by weathered polyethylene terephthalate and polyamide fibers. They hypothesized that these were not locally derived. The California Current carries waters south along the coast from British Columbia. Hence, a possible source might be microfibers released from wastewater treatment plants practicing primary-only treatment (see wastewater section) located north of Monterey.

In addition to biofouling, aggregation and incorporation into marine snow or fecal pellets can alter the buoyancy of the microplastic complex sufficiently to cause vertical export. Cole et al. (2016) tested the sinking rates of microplastic-containing copepod fecal pellets. Pellets with buoyant microplastics sank at a slower velocity than those without, making them more likely to fragment in the water column or be consumed by other organisms. In a laboratory test, Porter et al. (2018) found that microplastics were incorporated into marine snow, which increased their sinking rate. For polyamide fragments, this sinking rate increased by  $916\text{ m day}^{-1}$ . These authors also reported that mussels were more likely to ingest microplastics

incorporated into marine snow than bare microplastics. This demonstrated that marine snow is not only a route of vertical export but may also increase microplastic uptake by benthic organisms.

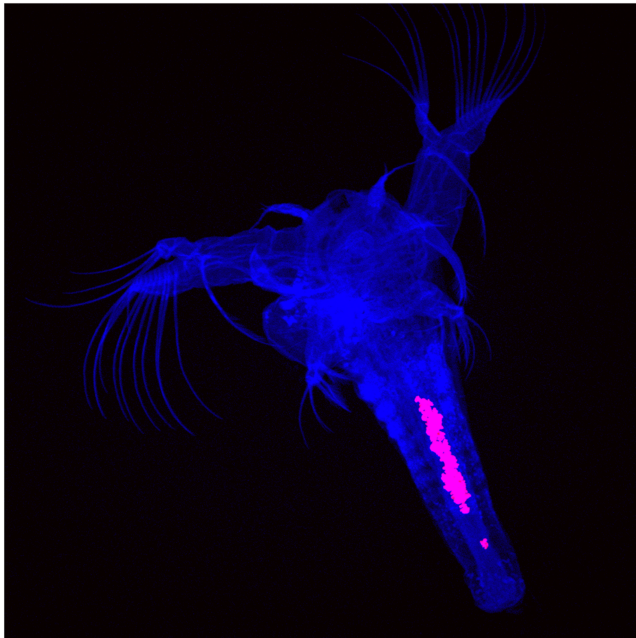
Microplastics are also abundant in coastal sediments, as evidenced by numerous surveys in coastal zones. Many studies have enumerated microplastics across sediments including beaches (Lots et al., 2017), shorelines (Browne et al., 2010), and coastal lagoons (Vianello et al., 2013). Van Cauwenberghe et al. (2015) reviewed studies of microplastics in coastal sediments, highlighting that most studies focused on beaches (80%). However, the sampling locations within beaches were highly variable between studies, reducing comparability. Furthermore, factors including wind and currents, transient conditions, and microplastic type complicate predicting locations of microplastic accumulation zones. Some studies, however, highlight that sediment suspension zones (associated with fine sediment particles) also have higher microplastic concentrations (Van Cauwenberghe et al., 2015; Vianello et al., 2013) than nonsuspension zones. As in coastal zone surface waters, microplastics in sediments appear to positively correlate with nearby population density (Browne et al., 2011; Van Cauwenberghe et al., 2015). Human population density also has a large influence on the type of microplastic found. Browne et al. (2011) reported high concentrations of microfibers near wastewater treatment effluent discharge sites, likely derived from textiles. Otherwise, microplastic fragments in coastal sediments are generally elevated in polyethylene and polypropylene, low-density polymers common in single-use plastics. However, denser polymers that are often in fiber form (e.g., polyester, polyamide, and acrylic) are regularly reported in high concentrations in intertidal and subtidal sediments, as well as sites with episodic polystyrene inputs (Erni-Cassola et al., 2019). Overall, these studies reveal that microplastics are abundant in coastal sediments, yet the trends are variable, limiting the efficacy of models based on coastal processes such as wind and currents alone. In total, although floating microplastics are the most commonly reported, multiple mechanisms exist that may remove even these buoyant particles from the surface ocean (Figure 5), thus transferring risks to other habitats, such as benthic ecosystems.

### 5.2.1. Biofouling

In aquatic environments plastics become covered by organic and inorganic materials, microorganisms, algae, and invertebrates. Zettler et al. (2013) coined the term “plastisphere” to describe this novel habitat. Biofilms on surfaces also increase drag on vessels. Development and manufacture of antifoulants to reduce growth on boat hulls, buoys, and other submerged surfaces is a major industry. The medical field is also concerned about the health consequences of microbial biofilm growth on teeth and surgical implants, such as heart valves and catheters. Biofilms serve as major avenues for human infections and potential mediators of antibiotic resistance (Socransky & Haffajee, 2002).

The biofilms that form on submerged surfaces in fresh and marine environments provide diverse niches, including aerobic and anaerobic zones, as well as pH, redox, and other gradients (Hall-Stoodley et al., 2004). Surface adhesion presents organisms with advantages, for example, greater nutritional access, physical protection, and environmental stability. Biofilm inhabitants also excrete chemical cues that affect invertebrate recruitment and forms of metabolic cooperation (e.g., quorum sensing; Socransky & Haffajee, 2002; Zhang et al., 2019). Further supporting the important ecological role of biofilms, Zhang et al. (2019) recently identified 7,300 new species from metagenomics data from biofilm-forming microorganisms. Most previous work to identify marine species focused on free-living forms. The additions of Zhang et al. (2019) increased the known microbial diversity of the oceans by more than 20%. Clearly, factors that influence or alter the formation and evolution of biofilm communities may have profound impacts on ecological communities in aquatic systems, in general.

Biofilms may also support habitats for microorganisms that excrete extracellular enzymes capable of degrading polymers (Dang & Lovell, 2016). Scanning electron microscopic examination of surfaces by Zettler et al. (2013) of weathered polyethylene and polypropylene debris from the North Atlantic revealed depressions, whose shapes conformed to those of the observed bacteria. They hypothesized that these depressions were caused by hydrocarbon degraders. Not surprisingly, they observed that communities on the plastic surfaces differed from those in the surrounding water. The authors also noted an abundance of *Vibrio* spp. on samples. This genus includes several pathogenic species (e.g., *V. cholera*, *V. vulnificus*, and *V. parahaemolyticus*) responsible for cholera, necrotizing wound infections, sepsis, and gastroenteritis in humans (Bartlett & Azam, 2005). In a substrate recruitment experiment in the North Sea, Oberbeckmann et al. (2016) used 16S rRNA gene sequence comparisons and reported that bacterial/archaeal communities associated with polyethylene terephthalate debris differed significantly from free-living ones, but not from those on



**Figure 6.** Image of polyurethane microplastics ( $<53\ \mu\text{m}$ ) ingested by brine shrimp nauplii (*Artemia sp.*, length  $\sim 500\ \mu\text{m}$ ). Microplastics were present at a concentration of  $100\ \text{mg L}^{-1}$ . Fluorescent microplastics (pink) are evident at a high density within the shrimp's digestive tract. These were egested within 48 hr after cessation of exposure. Some of the additives within the microplastics likely leached out of the plastic during its residence in the digestive tract and exposure water (see Figure 8). Imaged on an Olympus FV1200 laser scanning confocal microscope. Credit: Hamish Small (VIMS) and Virginia Worrell (Virginia Governor's School).

natural suspended particles ( $>3\ \mu\text{m}$ ) or glass. They did, however, observe differences at the operational taxonomic unit (OTU) level, with polyethylene terephthalate-associated communities exhibiting the presence of some hydrocarbon degraders. They also noted that biofilm communities differed by location and season.

The presence of biofilms may influence the likelihood of microplastic ingestion due to chemical cues exuded by the colonizing organisms. For example, in lab exposures Vroom et al. (2017) observed that polystyrene microplastics, primed by immersion in seawater, were ingested by two of three marine zooplankton species tested at a greater rate than unprimed plastics. Savoca et al. (2016) hypothesized that the odor of dimethyl sulfide-related chemicals that accumulated on plastics increased their consumption by sea birds. Savoca et al. (2017) later observed that aqueous leachates generated from weathered/biofouled polypropylene beads resulted in foraging behavior in the northern anchovy (*Engraulis mordax*) mimicking that observed when extracts of food were provided. Exposure to unfouled beads did not elicit such a response. Nasser and Lynch (2016) reported that proteins released from *Daphnia magna* (a common freshwater toxicity assay organism) associated with polystyrene nanobeads increased their agglomeration. Particles with these “eco-coronas” were ingested and retained at greater rates than untreated nanoparticles, and exposed organisms exhibited depressed feeding rates and lower survival.

Selective feeding on biofilmed, weathered plastics does not always occur, however. In a series of lab experiments, Allen et al. (2017) observed that a scleractinian coral ingested virtually all microplastic fragments (seven different polymer types of 500 to  $1,000\ \mu\text{m}$ ) offered but rejected similarly sized sand particles. In subsequent feeding experiments the corals ingested threefold to fivefold greater numbers of unweathered than fouled

plastics. The authors speculated that the biofilm masked chemical cues on the plastic that stimulated feeding (e.g., plastic additives), the physical weathering removed these cues or altered the surface properties, or the biofilm itself contained feeding deterrents. Nakashima et al. (2016) also hypothesized that biofilms forming on plastic debris might reduce the leaching of metal and organic additives, which may serve as sensory cues encouraging or discouraging ingestion. Holmes et al. (2012) observed that weathered polyethylene pellets adsorbed greater amounts of trace metals than virgin pellets. They hypothesized that this was due to an increase in surface reactivity derived from weathering, precipitates, or biofilms on the pellets. Clearly, the implications of biofilm formation vary across plastic composition and location and are likely perceived differently depending upon the species studied. Future research should investigate if plastic-degrading communities identified on microplastics in the field can facilitate identification of species or conditions capable of degrading polymers or aid in our design of new plastics that are readily degradable under real world conditions.

## 6. Microplastic Uptake and Consequences in Biota

As plastic debris is ubiquitous, essentially all aquatic and terrestrial species encounter it, regardless of trophic level. Biological effects may arise due to physical interactions with the microplastics, chemical exposure to plastic constituents (e.g. additives), sorbed toxins (synthetic or natural), or surface-associated organisms (e.g. pathogens). Interactions can lead to entanglement, ingestion, death, and a variety of other health or ecosystem effects. In the case of microplastic debris, the nature of the interactions is easily visualized (Hammer et al., 2012). Small aquatic biota, such as plankton, and larger filter feeders, from shellfish to whale sharks, may ingest or otherwise contact microplastic particles. Microplastic size, shape, and texture will be important in determining if there are negative consequences, yet this is not easily observed due to the small size and complex nature and behavior of microplastics. However, even minute zooplankton may ingest



microplastics (Cole et al., 2013). The physical impacts of microplastics on marine biota were reviewed by Wright et al. (2013). Figure 6 shows microplastics ( $<53\ \mu\text{m}$ ; produced from commercial polyurethane foam) ingested by brine shrimp nauplii (*Artemia sp.*; 400–500  $\mu\text{m}$  in length) in the lab. These microplastics were fluorescent due to the chemical additives present.

### 6.1. Microplastic Ingestion and Tissue Translocation

Exposure is a prerequisite to manifestation of toxic effects. Ingestion of microplastics by aquatic biota has been widely surveyed (Possatto et al., 2011; Lusher et al., 2013; Van Cauwenberghe & Janssen, 2014; Desforges et al., 2015; Güven et al., 2017; Lusher, Hollman, et al., 2017; Provencher et al., 2018; Nelms et al., 2019; Windsor et al., 2019). Marine species have been a focus of the bulk of such research. For example, Lusher et al. (2013) reported that 36% of pelagic and demersal fish collected from the English Channel had microplastics in their digestive systems. Nelms et al. (2019) observed that every specimen sampled of stranded marine mammals along the British coast exhibited microplastics in its digestive tract. In both studies, most were fibers, not fragments. Interactions with microplastic particles or fibers extend to lower trophic levels. For example, Van Cauwenberghe and Janssen (2014) reported that up to 50% of commercially grown bivalves contained microplastics in their tissues, and Desforges et al. (2015) observed microplastics (mostly fibers) in two zooplankton species collected from the Northeast Pacific. Choy et al. (2019) detected microplastics in giant larvaceans (*Bathochordaeus stygius*) and pelagic red crabs (*Pleuroncodes planipes*) at intermediate depths off the California coast. The larvaceans ingest particles using large mucous mesh filters, which are discarded when clogged, and then sink to the seafloor. While they did not focus on microplastics, Lamb et al. (2018) estimated that coral reefs from the Asia Pacific region were entangled with 11.1 billion plastic items. Rotjan et al. (2019) observed on average  $>100$  microplastics (predominantly polyamide, polyester, and synthetic cellulose-based fibers) per polyp of northern star coral (*Astrangia poculata*) collected off of Rhode Island (U.S.). In this same report corals coexposed in the lab to polyethylene microbeads and brine shrimp eggs of similar proportions preferentially ingested the microplastics. When fed microbeads initially, corals later failed to consume brine shrimp eggs. Seabirds have also been widely studied. Avery-Gomm et al. (2018) observed that 79% of sea bird (northern fulmar, *Fulmarus glacialis*) samples collected from the south-eastern Canadian waters of the Labrador Sea contained microplastics in their digestive tracts. Remarkably, Jamieson et al. (2019) recovered mainly microfibers from 72% of the *Lysianassoidea* amphipods sampled at 7,000 to 10,890 m in six of the deepest ocean trenches in the Pacific Rim. In freshwater environments, a recent study found that 50% of riverine macroinvertebrates (including detritivores and filter feeders) contained microplastics (Windsor et al., 2019). These and other studies provide proof that ingestion of microplastics by aquatic biota is widespread. Indeed, as the discard of plastics increases and our detection capabilities improve, percentages of occurrence will increase.

The motivation to ingest microplastics may be a combination of accidental ingestion and reaction to sensory cues but will vary by species, environment, and life stage. Cues may be visual, such as color or shape (Schuyler et al., 2014) or tactile. As discussed above, plastic additives or cues emitted from associated biofilms may influence ingestion. Terrestrial wildlife, for example, bears and wolves, have also been observed to ingest plastic marine debris on Alaskan shores. Microplastics were apparent in their feces and tooth and claw marks observed on larger debris (Figure 7). This demonstrates another fragmentation process, as well as the transfer of plastics from marine to terrestrial ecosystems. In a freshwater to terrestrial transfer context, Windsor et al. (2019) observed microplastics in half of the freshwater feeding invertebrates sampled, including mayflies and caddisflies. In the lab, Al-Jaibachi et al. (2018) showed that microplastics could be transferred between aquatic (larvae) and terrestrial (adult) stages of mosquitos (*Culex pipiens*). Small particles (2  $\mu\text{m}$ ) were more efficiently transferred than larger (15  $\mu\text{m}$ ) polystyrene beads.

There is compelling evidence that microplastics  $<20\ \mu\text{m}$  may enter the tissues of diverse biota through the digestive system, hemolymph, or blood. Studies have shown that microplastics translocate in the tissues of invertebrates such as fiddler crabs and mussels (Brennecke et al., 2015; Browne et al., 2008). Dawson et al. (2018) fed  $\sim 30\ \mu\text{m}$  fluorescent polyethylene microbeads to Antarctic krill (*Euphausia superba*), the major phytoplankton grazer in the Southern Ocean. They observed that fragments isolated from the krill and their feces were on average 6 to 7  $\mu\text{m}$ , while fragments  $<1\ \mu\text{m}$  entered the digestive gland itself. The authors suggested that in nature, particles will weather and become embrittled, increasing vulnerability to physical breakdown during digestion. Such fragmentation to microplastics and nanoplastics may facilitate their



**Figure 7.** Fragmentation and ingestion of marine plastic debris by Alaskan terrestrial wildlife. (a) Plastics washed up on Alaskan beach, including tsunami debris; (b) polystyrene fragmented by wildlife; (c) PVC float chewed on by wildlife; (d) polystyrene floats chewed on by wildlife; (e) bear scat, showing ingested plastic debris; (f) coyote scat containing ingested plastic debris. Photos courtesy of Chris Pallister, Gulf of Alaska Keepers (goak.org).

translocation into tissues. Investigation of several finfishes showed that microplastics accumulate in the gills, alimentary tract, liver, and muscle (Avio et al., 2015; Choi et al., 2018; Greven et al., 2016; Su et al., 2019), with distributions among organs differing depending on particle size (Lu et al., 2016). Ding et al. (2018) exposed red tilapia fish over 14 days to  $0.1 \mu\text{m}$  polystyrene particles at 1, 10, and  $100 \mu\text{g L}^{-1}$ . Tissue concentrations increased over time in the order: alimentary tract > gills > liver/brain). Transport across the highly selective blood brain barrier and accumulation of nanoplastics in the brain of fishes indicated that small microplastics may enter the vascular system and be transported to diverse internal tissues, including blood filtering (e.g., liver) and immune organs (e.g., spleen and head kidney). Entry of microplastics and nanoplastics into tissues may also facilitate food chain transfer. Mattsson et al. (2017) found that nanoplastic particles (52–330 nm) were transferred up through a marine food chain, from zooplankton to finfish.

Ingestion or tissue translocation of microplastics can cause digestive system blockage, tissue damage, behavioral changes, immune response, and death. Yet, some encounters may result in no observed effects (Jovanović, 2017). Outcomes are dependent on the type of debris and organism and can be highly variable. Mortality is generally not a dominant observation. More commonly, microplastic ingestion is reported to reduce consumption of nutritious prey, leading to altered metabolism and behavior (as summarized in Galloway et al., 2017). Contrarily, an experiment where brine shrimp larvae were exposed to  $10 \mu\text{m}$

polystyrene particles found no effect on development, growth, or survival but reported that epithelial tissues of the intestinal tract were changed, possibly affecting digestive efficiency (Y. Wang, Zhang, et al., 2019). Similarly, Pedà et al. (2016) observed damage to the intestine of European sea bass following polyvinyl chloride pellet (3 mm) exposure and Jin et al. (2018) found that the intestinal epithelial tissues and mucous of mice were altered following ingestion of 5  $\mu\text{m}$  polystyrene particles during a 6-week exposure. These observations suggest that a major consequence of ingestion of microplastics may be filling or physical alteration of the digestive tract, compromising ingestion or assimilation of nutritious food.

## 6.2. Toxic Effects of Microplastics

Behavioral, metabolic, and developmental changes have been observed following microplastic exposure. Mattsson et al. (2017) found that nanoplastic particles (52–330 nm) reduced survival of zooplankton and altered behavior in finfish (measured as activity, feeding time, and distance swam in search of prey). Although few studies have investigated effects across multiple life stages, Luan et al. (2019) observed that polystyrene microplastics were most toxic to developing clams during the egg hatching stage (compared to larval stages), decreasing developmental rates. Research has also shown that microplastics may disrupt development leading to physical abnormalities, such as Nobre et al. (2015) reported in sea urchin embryos. Oxidative stress has also been observed following exposure in a variety of species, including the aforementioned reports on juvenile zebra fish (Lu et al., 2016) and red tilapia (Ding et al., 2018). Overall, a variety of studies have addressed these effects (reviewed in Prokić et al., 2019). However, the diversity of microplastic materials and metrics of effects used make it problematic to directly compare results, just as the diversity of microplastics in the natural environment makes it difficult to predict toxicological outcomes with confidence.

The innate and adaptive immune systems are designed to respond to foreign antigens, so research has queried if microplastics may be perceived as antigens or possibly inhibit immune response. Espinosa et al. (2018) studied immune responses of head kidney leucocytes in seabream (*Sparus aurata*) and sea bass (*Dicentrarchus labrax*) using 40–150  $\mu\text{m}$  polyethylene and polyvinyl chloride particles. They found that there was a minimal immune response, with the exception of potential oxidative stress. However, this study used microplastics 4 to 15 times larger than the average immune cell. As such, direct effects on function of immune cells (including phagocytosis) would be limited. Veneman et al. (2017) tested developmental effects of polystyrene (0.7  $\mu\text{m}$ ) in early stage zebra fish. The authors noted interactions with phagocytic cells (including neutrophils) and activation of the complement system and neutrophils. Greven et al. (2016) observed changes in fathead minnow neutrophil response after exposure to polystyrene and polycarbonate nanoplastics. These and other studies suggest that microplastics can induce immune responses in aquatic species, notably fishes (whose immune system is a proxy for human immunity), which may alter infectious disease resistance.

Recent work has investigated the influence of microplastics on organismal microbiomes. The importance of microbiomes to nutrition and disease protection is well recognized in fishes (Adamovsky et al., 2018; Llewellyn et al., 2014) and humans (Shreiner et al., 2015). As previously discussed, microplastic surfaces may develop novel biofilm microbiomes, potentially leading to specific microbial-mediated consequences (Zettler et al., 2013). Jin et al. (2018) found that in addition to digestive tissue alterations, the gut microbiome of mice was substantially altered following polystyrene microplastic ingestion, modifying metabolic pathways including amino acid and bile metabolism. These authors suggested that such alterations could trigger metabolic disorders. Wan et al. (2019) monitored the microbiome of larval zebrafish after 1 week of exposure to 5 and 50  $\mu\text{m}$  polystyrene particles (at 100 and 1,000  $\mu\text{g L}^{-1}$ ). During this time, microbial diversity and metabolic functions (i.e., lipid metabolism and glycolysis) were significantly altered. In wild-caught flesh-footed shearwaters (*Ardenna carneipes*), an endangered seabird in Australia, Lavers et al. (2019) observed a correlation between blood chemistry parameters (lowered blood calcium, heightened uric acid, cholesterol, and amylase) and reduced morphometrics (reduced body mass, wing length, culmen, and head + bill length) with the incidence of ingested plastic.

Research has also demonstrated that reproduction may be affected by microplastic exposure. A study investigating the effects of polystyrene microbeads (2 and 6  $\mu\text{m}$  at 32  $\mu\text{g L}^{-1}$ ) found that mussels exhibited decreased reproductive capacity following a 2-month exposure (Sussarellu et al., 2016). This was evidenced in the abundance and development of larvae, as well as the number and diameter of oocytes and sperm motility. The authors suggested that this resulted from an energy shift away from reproduction and toward structural growth, due to feeding changes triggered by microplastic exposure. In a similar study with *Daphnia*



*magna*, individuals fed 1–5  $\mu\text{m}$  microplastic particles over a 21-day incubation exhibited decreased reproductive success (Pacheco et al., 2018). Effects included delayed brood release, decreased number of brood, and immobility in juveniles.

Most of the available toxicological studies have been conducted under laboratory conditions, which may limit their real-world applicability. Many used fluorescently labeled plastics to track their distribution through tissues (e.g., Browne et al., 2008; Choi et al., 2018; Ding et al., 2018; Dawson et al., 2018; Greven et al., 2016; Jin et al., 2018; Lu et al., 2016; Veneman et al., 2017; Wan et al., 2019; Y Wang, Zhang, et al., 2019). Toxicologists must ensure that the dyes do not leach or possess toxic properties. Schür et al. (2019) observed that the fluorescence signal does not always indicate particle location itself but may be an artifact of fluorescent chemical translocation. A reason that few studies have addressed toxicity in field-collected scenarios is that unlabeled (e.g., nonfluorescing) microplastics in such samples are difficult to detect within organisms. Although research has specifically addressed methodologies used to separate microplastics from tissues (Lusher, Welden, et al., 2017), microplastics  $<20\ \mu\text{m}$  are difficult to isolate and identify. In addition, microplastics in the environment are extremely diverse, that is, exist as complex mixtures, in a sea of additional potential stressors. Thus, tying effects to a single, particular plastic can be extremely difficult outside a controlled, laboratory situation. Clearly, further research on the dissemination of microplastics into and between tissues is necessary to elucidate toxicological consequences. A major challenge to this research is increasing our ability to reliably trace small microplastics.

### 6.3. Mechanisms of Microplastic Toxicity

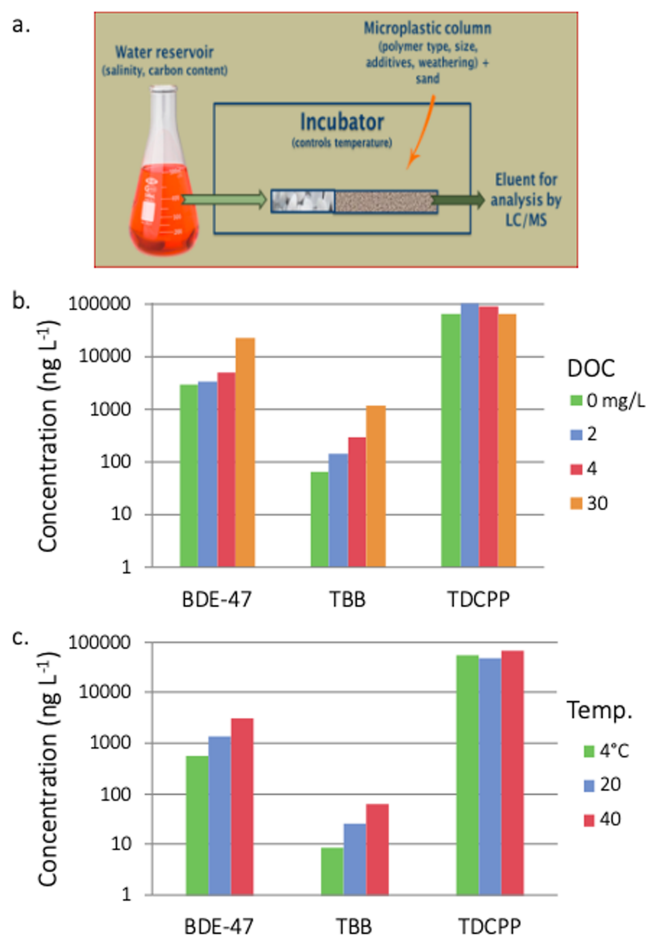
Pathologies due to microplastic exposure can be driven by a variety of factors, including sorbed pathogens/pollutants and polymer/additive composition. As previously discussed, it has been demonstrated that microplastics can harbor pathogens (Kirstein et al., 2016; Viršek et al., 2017). Plastic/biofilm associations have been proposed as a vector of disease in wildlife and humans (Keswani et al., 2016). Biofilms on plastics used in human joint replacements have been reported to be a source of infection (Gbejuade et al., 2015). Lamb et al. (2018) observed disease incidence increased from 4% to 89% when coral reef species were in contact with plastics. Rotjan et al. (2019) fed corals in the lab polyethylene microplastics with an *Escherichia coli*-containing biofilm. Two weeks postingestion *E. coli* was detected in all such fed corals, as well as in some adjacent polyps. These corals died within 4 weeks after microplastic ingestion.

Microplastics sorb persistent organic and inorganic (e.g. toxic metals) pollutants in natural environments. Indeed, this phenomenon has been exploited in designing polymer-based passive sampling devices for dissolved organic pollutants (MacKenzie et al., 2004; Rochman et al., 2013; Telfer et al., 2014). However, sorption of a pollutant to a microplastic surface may be influenced by competitive interactions with other chemicals present, as demonstrated by Bakir et al. (2012). The exaggerated surface areas of microplastics compared to parent debris enhance contaminant sorption potential. For example, J. Wang, Liu, et al. (2019) examined the sorption of phenanthrene and nitrobenzene onto polystyrene microplastics. Particle/water partitioning of these chemicals tracked decreasing particle size, except for the smallest nanoparticles where aggregation may have reduced their cumulative surface areas. Both laboratory (Beckingham & Ghosh, 2017; Tanaka et al., 2015) and field-based studies (Gassel et al., 2013; Rochman et al., 2014; Scopetani et al., 2018) investigated whether such sorbed pollutants accumulate in organisms after ingestion. Batel et al. (2016) fed *Artemia* sp. nauplii small microplastics with absorbed benzo(a)pyrene. They then fed these nauplii to zebrafish (*Danio rerio*) and observed trophic transfer of both the microplastics and benzo(a)pyrene. Most such studies examined organic pollutants, but Barboza et al. (2018) evaluated interactions between mercury and microplastics, and resulting toxicological implications in fish.

Some authors (e.g., Besseling et al., 2018; Koelmans et al., 2016) have suggested that the importance of contaminant accumulation potential may be overestimated. They hypothesized that the outcome varies depending on the conditions within the gut of the organism studied, that is, the type of gut fluid (Tanaka et al., 2015). Contaminant sorption onto plastics could also reduce the opportunity for contact with the organism, for example, if the microplastic/contaminant is sequestered in sediments.

Plastic products also may contain toxic organic and inorganic additives at several percent of the total plastic weight (Hermabessiere et al., 2017). Plastics are widely used as containers for food and beverages, as well as in medical devices, and researchers have examined additive migration in those contexts (Hahladakis et al., 2018), but few have considered the toxicological potential of microplastic-related additives on aquatic life.





**Figure 8.** Leaching of additives from polyurethane microplastics under different environmental conditions. (a) Microplastics were added to a sand column, leachate collected, and then analyzed by liquid chromatography/mass spectrometry (LC/MS). Increasing (b) dissolved organic carbon (DOC) concentration and (c) temperature greatly enhanced release of the hydrophobic BDE-47 and TBB, but not the more hydrophilic TDCPP. [BDE-47: 2,2',4,4'-tetra-bromodiphenyl ether; TBB: 2-ethyl-hexyl tetrabromobenzate; TDCPP: tris(1,3-dichloro-2-propyl)phosphate. Default leaching conditions: 400 mg of 53–300  $\mu$ m polyurethane, water column flow 1 ml/min, 40 °C, 0 PSU, no added DOC.]

Lead, an additive in some polyvinyl chloride fishing floats, was shown to leach in lab experiments (Nakashima et al., 2016). Migration decreased over time as the lead near the float's surface was depleted. However, release was reinvigorated after the polymer surface was abraded, as might occur as the plastic fragments in the field. Thus, organisms contacting virgin microplastics may experience greater exposure to additives than those encountering weathered materials (Nobre et al., 2015).

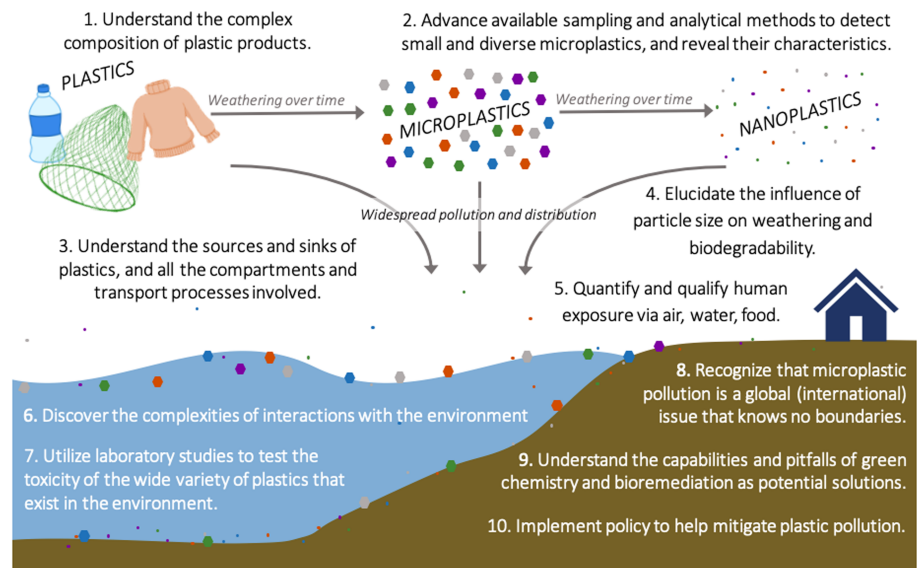
The characteristics of the polymer, additive, and the leaching fluid play a role in the extent of the additive release. For example, Hale and Chen (2016) examined the leaching of flame retardants from virgin polyurethane microplastics in the laboratory under various salinity, temperature, DOC, and digestive fluid conditions. Increasing water temperatures enhanced the release of hydrophobic brominated flame retardants, but not the more soluble phosphate-based additives, which was already substantial. Increasing humic acid concentrations showed a similar pattern (see Figure 8), as did surrogate digestive fluids (not shown). As there typically are multiple sources of additives (e.g., wastewater treatment plant effluents, industrial releases, and contaminated sediments), the contribution of leaching directly from in situ plastic debris is difficult to assess. However, Martins et al. (2015, 2016) reported phthalate uptake and plastic debris-derived immunogenic effects in salmonid fishes in a remote Alaskan estuary. The site was pristine except for input of ocean-derived plastic debris.

In some cases, the presence of specific additives in plastics is unanticipated. For example, polystyrene fishing floats, aquaculture buoys have been observed to contain the flame retardant hexabromocyclododecane (HBCD; Rani et al., 2014; Jang et al., 2016) and in shellfish growing thereon (Jang et al., 2016). While HBCD has been widely used as an additive in extruded polystyrene insulation boards, there is no functional reason for it being in floats. The authors observed some of the highest HBCD concentrations in polystyrene insulation board debris from the Pacific coastlines of Alaska and California (Jang et al., 2017), theorized to be from the 2011 Japanese tsunami disaster.

In a terrestrial context, Gaylor et al. (2012) observed that house crickets (*A. domesticus*) ingested polyurethane microplastics and accumulated a portion of the polymer additives therein. Previous work had shown that such additives could undergo trophic transfer to frogs via consumption

of polyurethane-exposed crickets (Hale et al., 2002). These same polymer additives had also been detected at high levels in fish collected from a river downstream of a treatment plant (Hale et al., 2001) that received wastewater from a plastics manufacturing facility. However, the precise role played by microplastics in transferring the additives to the fish has yet to be delineated. Nonetheless, polymer additive detection in fish fillets confirmed their presence outside of the digestive tract and in tissue consumed by humans.

Toxicity identification evaluation approaches may advance our understanding of the extent and consequences of polymer additive and sorbed contaminant release. In this context, Coffin et al. (2019) examined the ability of pepsin, a digestive enzyme, to leach additives from common plastic items. Using an in-vitro cell line, they reported that simulated gut conditions increased the estrogenicity of leachates from polystyrene, polyethylene, and polypropylene. Bisphenol A and two phthalate plasticizers were identified, but levels detected were deemed insufficient to explain the estrogenicity observed. The authors hypothesized that additional, undetermined additives in the leachates likely contributed. Jonker et al. (2016) combined chemical analysis (liquid chromatography/high-resolution time-of-flight mass spectrometry) of plastic leachates with estrogenicity detection. They employed a reporter gene assay incorporating human VM7Luc4E2 cells. The plastics were extracted with organic solvents (tetrahydrofuran:methanol; 50:50, v/v), which revealed



**Figure 9.** Challenges for the study and reduction of microplastic pollution are illustrated here. Note that microplastics and nanoplastics have been simplistically illustrated here as hexagonal, colored particles. In actuality, their sizes and shapes are extremely variable and include fibers. Challenges 1–4 stem from the nature of the plastics and their weathering with time. Challenges 5–7 involve determining their distribution throughout the environment results and delineation of their effects. Finally, Challenges 8–10 involve mitigating the global health risks that microplastics pose.

several brominated and phosphate-based additives. Estrogenic compounds identified included bisphenol A and 2,4-di-tert-butylphenol. The authors hypothesized that the lack of associated estrogenicity may have been due to the additives' low known potencies and water solubilities.

Plastic polymers themselves are generally viewed as inert, nontoxic, stable materials; however, exceptions exist. For example, the polystyrene monomer styrene has carcinogenic properties (Gibbs & Mulligan, 1997). Saido et al. (2014) found styrene to be widely distributed along coastlines of the northwest Pacific Ocean. Comparison of and extrapolation from lab studies using the same nominal polymer type can be misleading as materials obtained from different sources may differ in regards to actual polymer composition, morphology, and additive content. Composition of “neat” plastics obtained from commercial sources may differ from those commonly in consumer goods and thus in environmental debris. Furthermore, important compositional information may be deemed confidential business information and thus unavailable to the researcher.

Microplastic morphological characteristics (shape, texture, and size) may influence toxicological outcomes, for example, controlling if it is translocated in the tissues, blocks the digestive tract, or irritates or lacerates tissues, resulting in abnormalities or increasing susceptibility to infectious diseases. Growing research suggests that microfibers are a prevalent form of plastic pollution in the environment (Güven et al., 2017; Panno et al., 2019; Michielssen et al., 2016). Their dimensions may present novel risks, as they are more likely to penetrate or otherwise irritate tissues. The toxicity of asbestos, for example, is driven by its fibrous morphology (Siegrist & Wylie, 1980). Further, weathering changes the surface morphology and properties of microplastics (Balakrishnan et al., 2019) and may have profound effects on toxicological outcomes. For simplicity and reproducibility, most toxicological studies have utilized virgin plastics, although exceptions exist (see biofilm section). Standardized methods to realistically weather plastics prior to experimentation are needed. A more complete understanding of toxic outcomes and ecological risks from exposure to microplastics would justify increased expenditures to delineate their fate and reduce environmental releases, relative to the threats posed by other stressors such as disease, climate change and toxic chemicals (Koelmans et al., 2017).

## 7. Conclusions

Recently, concern over microplastics in the environment has been criticized as a distraction from other issues threatening global environmental health, including climate change (Stafford & Jones, 2019).

Ironically, plastics also play a role in climate change. Their manufacture represents 6% of current global oil consumption (Ellen MacArthur Foundation, McKinsey and Company, 2016). Zheng and Suh (2019) calculated that fossil fuel-based plastics produced in 2015 emitted 3.8% of the total global CO<sub>2</sub> over the course of their life cycles. The growth in plastic production is outpacing carbon emissions (Figure 1) and if unchecked is projected to contribute 15% of global greenhouse gases by 2050 (Ellen MacArthur Foundation, McKinsey and Company, 2016). Over 90% arose during plastic production versus end of life (landfilling, recycling, and incineration). Recycling would constitute a net CO<sub>2</sub> loss if used to supplant virgin plastic manufacturing (Ellen MacArthur Foundation, McKinsey and Company, 2016). Furthermore, weathering of plastics in the surface ocean has been shown to release the potent greenhouse gases methane and ethylene; thus, plastic is not a permanent CO<sub>2</sub> sink (Royer et al., 2018). Zheng and Suh (2019) suggested that increased use of renewable bio-based plastic feedstocks, greater recycling, and management of increasing demand are critical. However, Reddy et al. (2013) noted that a shift to generate 250 million tons of plastics from bio-based sources would divert 5% of available arable land from food production. In addition, the increased supply of natural gas from hydrofracturing of North American shale deposits has fueled an expansion in U.S. plastics manufacturing capacity (American Chemical Council, 2015). This poses an economic hurdle to increased recycling. Clearly, the environmental role of plastics is complex, and solutions will require creative strategies.

Due to the useful properties of plastics and our growing dependence on them, we cannot simply ban or quickly replace them. Indeed, global usage is escalating, as is their mismanagement and entry into the natural environment. These plastics will over time degrade into microplastics and nanoplastics. At present, we do not adequately understand the toxicological or ecosystem consequences of these. However, if serious impacts are not already occurring, they certainly will arise as environmental levels increase. Examining “hot spots” and the most exposed species are two valuable strategies for evaluating toxicological severity of pollution. Ironically, humans living indoors, not marine organisms, may be the most exposed to microplastics and associated additives.

This review is based on two underlying principles: (1) Microplastics are a transitory state existing between plastic products/macrob debris and nanoplastics. To understand and solve the “microplastics” issue, we cannot consider microplastics in isolation; we must consider all size groups. (2) The “microplastics” problem extends well beyond the oceans. While the ultimate sink of microplastics is likely deep ocean sediments, plastics are produced, used, and discarded initially on land and then are dispersed through the other compartments.

Grand challenges related to these are detailed below and summarized in Figure 9.

1. *Understand the complex composition of plastic products.* The perception that all plastics are inert and compositionally identical is incorrect. To resolve critical questions and mitigate possible impacts, plastic manufacturers, aquatic and terrestrial and atmospheric scientists, health care specialists, waste and chemical engineers, economists, regulators, and others must collaborate and better understand the composition and nature of plastic products, including additives. The complexity of microplastics becomes far more convoluted once they enter, intermingle, and weather in the environment.
2. *Advance available sampling and analytical methods to detect small (<20  $\mu\text{m}$ ) and diverse microplastics and reveal their characteristics.* We must improve sampling and analysis capabilities across all matrices, including nanoplastics and weathered materials. Failure to sample appropriate locales in a representative manner or to encompass critical analytes leads to invalid conclusions. An overemphasis on the abundance of microplastic particles in samples, in lieu of mass balances, currently exists. In part, this is driven by analytical procedures that are weak in terms of mass quantitation.
3. *Understand the sources and sinks of plastics, compartments, and transport processes involved.* With adequate analytical tools in hand, we can better elucidate the sources, pathways, and sinks of macroplastics, microplastics, and nanoplastics in terrestrial, aquatic, and atmospheric compartments. Estimates based on selected strata (e.g., surface waters or sediments), size ranges (e.g., sampling >300  $\mu\text{m}$ ), polymer types (e.g., confined to buoyant olefins), or forms (pellets, fragments, or fibers) should be qualified to avoid confusion. In addition, an oft-mentioned criticism of the sparse published toxicity studies available is that some are conducted at “unrealistically high microplastic concentrations.” Our current analytical capabilities are inadequate to determine true environmental concentrations and levels are increasing.

4. *Elucidate the influence of particle size on weathering and biodegradability.* Do particles/fibers continue to fragment into nanomaterials? Does the fate of these minute materials differ from expectations? That is, do plastics last indefinitely in the environment or do they become increasingly vulnerable to physical and biologically mediated degradation as they fragment and their surface areas increase (Figure 2)? This has been inadequately explored to date.
5. *Quantify human exposure via air, water, and food.* We are exposed to microplastics and nanoplastics via contaminated food and water. Ironically, much of this (and indoor air) is in intimate contact with plastic containers, filters, and other devices. We are likely most exposed to microplastics and associated additives from our indoor environment, yet little research to date has investigated this, let alone evaluated associated risks. It is instructive that ingestion and inhalation of indoor dust is now accepted as the major pathway for our exposure to flame retardant polymer additives. What about exposure to microplastics themselves, which are carriers of these additives?
6. *Discover the complexities of microplastic interactions with the environment.* We must better understand the complexities of microplastic interactions with the environment, especially transition zones such as estuaries. These must include holistic studies on individual organism, populations, and ecosystem health and processes. Although many studies have begun to elucidate microplastic fate, more are needed to prioritize prevention and remediation strategies. In addition, plastic surfaces submerged in aquatic systems rapidly acquire biofilms (“eco-coronas”) and may be chemically altered by, for example, oxidation. These affect biological ingestion/palatability, degradability, specific gravity, sorption of contaminants, and desorption of additives. Therefore, these environmental interactions should be carefully evaluated.
7. *Utilize laboratory studies to evaluate the toxicity of the wide variety of plastics that exist in the environment.* Controlled laboratory studies are critical in elucidating the effects of different plastic products on species and communities of interest. Such studies must be designed to address true, environmental threats. For example, many utilize plastic beads purchased from laboratory suppliers. Although these provide insights into important processes, they differ in form and composition from plastic debris in the environment, limiting the unrestrained applicability of results.
8. *Recognize that microplastic pollution is a global (international) issue that does not respect political boundaries.* Like the trajectories of climate change, fossil fuel consumption, and species management, developed and emerging nations must cooperate to find equitable solutions to plastics pollution.
9. *Understand the capabilities and pitfalls of green chemistry and bioremediation as potential solutions.* The most successful solutions to microplastic pollution will take place at the beginning of the lifecycle of plastic products themselves, not remediating microplastics once in the open ocean. This is similar to controlling or cleaning up an oil spill. Prevention is best, followed by containment and lastly cleanup. The development of “green materials” is promising. However, facile breakdown of the polymer matrix may facilitate release and enhance exposure to potentially toxic additives, so additives must also be thoughtfully engineered—we must avoid the so-called “regrettable substitution.” Additionally, plastic waste management and recycling must be improved. Developing countries have recently been identified as an increasing source of marine plastic pollution. Wealthy nations should pioneer strategies for redesign, reuse, and recycling, not seek to offload their wastes to developing nations.
10. *Implement policy to help mitigate plastic pollution.* Most plastics are inexpensive to manufacture. Hence, there is little financial incentive to reuse them. To support a circular lifecycle, the upfront price of plastics must incorporate end of life costs. Currently, low volume plastic users and associated ecosystems bear a disproportionate burden (e.g., remote islands are now being littered with plastic debris). This environmental injustice echoes that of climate change and sea level rise. Landfills may be mined by future generations as resources become scarce and technologies improve. Optimization of such “dumps” into “repositories” is worthy of consideration. Political initiatives across borders should seek to accomplish these goals.

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