Source tracking microplastics in the freshwater environment

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ABSTRACT

Plastics are a frequently observed component of marine debris and there is growing concern about microplastic (MP) ecotoxicity, and the impacts of additives, sorbed hazardous organic contaminants, heavy metals, and biofilm on MP surfaces. The relative importance of MP from different terrestrial and freshwater sources is poorly understood and limits our ability to develop best management practices. This review focuses on evidence and methods for source apportionment of MP in freshwater environments including the use of MP characteristics, mass balance techniques, and surface characteristics. Within-study data indicated some potential for differences in polymer identity and morphology for differentiating select sources, but clear cross-study patterns were lacking. Major challenges identified include technical challenges in accurately identifying polymers, multiple classification schemes for reported MP morphologies, lack of data for several terrestrial sources, poor understanding of differential fate/transport/weathering processes for MP and surface contaminants, and methodological difficulties simultaneously confirming polymers and surface contaminants.

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1. Introduction

Research conducted on occurrence of environmental microplastics (MP) has primarily focused on determining MP abundance in marine environments. Studies have been conducted to determine the prevalence of MP on the sea surface, within the water column, and in marine sediments along beaches. A review of 68 research articles conducted by Hidalgo-Ruz et al. [1] found microplastic particles ranged from 1 μm to 29 mm with concentrations varying by 7 orders of magnitude within all three of these compartments. Comparatively less research has been done on the abundance of MP in the freshwater environment; however, the growing number of studies that have been conducted offer strong evidence to support the existence of MP contamination in freshwater systems [2]. A recent review by Li et al. indicated that quantification of the MP particles in freshwater range from greater than 1 million pieces per cubic meter to less than 1 piece in 100 cubic meters [2]. A better understanding of the wide variety of primary (manufactured in small sizes) and secondary (derived from the breakdown of other macro and micro-plastic) MP sources in freshwater is needed.

MP are generally defined as polymer particles <5 mm in size that present in a variety of morphologies (i.e., beads, fragments, fibers, films). Determining the source of MP is complicated by the fact that these particles can travel far from their point of origin, evidenced by observations of MP in remote regions of the world (e.g., Ref. [3]). There is also potential for different fate (e.g., biological uptake, photodegradation, biofouling and settling) and transport processes that may alter the profile of MP observed at a source compared to what may be observed at a sampling site that could complicate fingerprinting approaches to source tracking if not accounted for.

Mitigating MP pollution in the freshwater environment requires understanding the sources and their relative importance in terms not only of quantity but also of any potentially adverse impacts and environmental fate and transport. The implementation of voluntary phaseouts and legislation banning the addition of plastic bags and MP particles to personal care products is an example of how mitigation strategies can be implemented when sources are well defined (in addition to there being other circumstances that facilitate change) [4,5]. Multiple global reports [5,6] agree that a
The lack of consistent methodologies for identifying polymers may significantly improve the ability to analyze more samples within a reasonable time frame is desirable and development is on-going.

A small number of studies have inferred the source of MP contamination based on the polymer. Zhang et al. [19] determined that polystyrene identified in the Bohai Sea in China was likely due to floating foam rafts from marine agricultural areas and Styrofoam food containers, both of which were observed in a reconnaissance of beaches and shoreline near the sampling areas. Similarly, Xiong et al. [20] identified the dominant polymers detected in water samples to be polyethylene, which was attributed by the authors to improper waste disposal of food packaging and containers along the shoreline. In contrast to observed differences in MP morphology, the polymer profile was not reported to be significantly different between influent and effluent samples in a 10 treatment plant study in the Netherlands [10]. Browne et al. [21] identified a predominance of polyester and acrylic fibers in shoreline sediments as well as wastewater effluents, suggesting that wastewater inputs were the predominant source of fibers at these locations.

Fig. 1 provides an analysis of reported polymer composition across 35 wastewater treatment plant effluent samples, one wastewater influent sample, one grit sample, one grease sample, one sludge sample, one wastewater solids sample and six compost samples. It includes data from four studies published over a two-year period. Taken together, over 30 different polymers were identified in these samples. The composition of wastewater effluent samples was highly varied with some effluent samples containing only 2 polymers, while others contained up to twelve different polymer types. Similarly, the compost samples contained ≥4 polymers with no consistent trends observed in the percentage of a given polymer among the samples.

The morphology of MPs may also provide some evidence of their source [7]. The presence of primary MP morphologies (microbeads from personal care products and fibers from laundering of textiles) are likely associated with wastewater inputs [22]. Similarly, secondary MP such as films and fragments are associated with weathering of plastic packaging or plastic bags and may be associated with terrestrial sources such as improper solid waste disposal. Another source of MP fragments is stormwater runoff. Horton et al. [23] identified a storm drain carrying urban roadway runoff directly upgradient of a sampling location as the source of the increased abundance of fragments identified at this site.

We analyzed nineteen paired wastewater influent and effluent samples originally reported by Magnusson and Wahlberg [24] and Michaelsen et al. [25] and identified the primary morphologies in each sample (Fig. 2a). The primary morphologies present in each of the paired samples were fibers, films and fragments. Morphology relative percent differences ranged from 11 to 147% for paired influent and effluent samples with an average of 85 ± 41% relative percent difference for the two sample types. When comparing the paired influent and effluent from each location, there were decreasing percentages of fibers and films in the effluent as
compared to the influent in 10 and 8 samples, respectively. In contrast, the fragment morphology increased from influent to effluent in 12 of the 19 samples. This indicates that fibers and films are more easily removed in wastewater treatment unit processes and may be more likely to be associated with municipal biosolids. These morphologies may enter freshwater systems predominantly through surface runoff after land application of biosolids, while fragments may be more commonly associated with wastewater effluent discharges into receiving streams. Combining morphological data across studies and matrices (water, biosolids) and levels of treatment (influent, effluent) did not indicate clustering by source (Fig. 2b). Here, we applied ordination [non-metric multidimensional scaling (nMDS)] of square root transformed percent abundance data for the different reported MP morphologies for different studies and clear clustering was not observed. However, this does not necessarily mean morphology is not a useful indicator: it should be noted that cross study comparisons are challenging given that the shape classes reported vary and may include fragments, beads, fibers, pellets, paint chips, films, and foams, and that the morphologies observed for a given sample can vary by the size class targeted. It is also possible that these patterns are not consistent across geographies.

3. Source sampling and mass balance to establish inputs

Direct sampling of aqueous and solid waste streams has been used to characterize the sources of MP pollution in freshwater systems. A major focus of these studies has been understanding the impact of municipal wastewater effluent as a pathway for MP entry into surface water [10–12,24–30]. Many of these studies include calculations for daily emissions of MP to the receiving water bodies (or these values are easily calculated where average daily flow rates are reported). Comparing emission mass fluxes (emission concentrations × flow rates) to the receiving water body mass fluxes can inform the relative importance of wastewater effluent as a point source of MP, although these dilution factors are rarely reported. Some researchers did compare the MP concentration in wastewater effluent to surrounding surface water, generally implicating the effluent as a source based on its higher concentration [26,27,29]. Studies have also sought to compare up and downstream MP concentrations to implicate wastewater effluent as a point source [31], in this case the majority of the downstream sampling sites did not have significantly higher MP concentrations compared to the paired upstream sites. In the case of sanitary and combined sewer overflows, wastewater influent would be released to freshwater sources and influent generally had an average (per study) of 63–420 times higher concentrations of MP (Fig. 3). Variation in the total MP concentration for different studies is likely due to some combination of differences in target MP size class and other analysis techniques, study site, temporal variation, and whether MP were identified visually (considered prone to false positives and negatives) [32] or with confirmation of chemical structure. The MP composition in wastewater influent would be expected to be impacted both by the inputs and activities in the sewershed (i.e., fibers from laundry, industrial inputs if present, stormwater if combined sewers, use of consumer products, etc.) and the treatment performed at the facilities themselves that can impact the effluent MP fingerprint and concentration. Differentiating these sources contributing to wastewater influent is complex and system specific. The best information available is regarding the contribution of the laundry fraction from households (as reviewed by Ref. [33]).

Runoff containing MP from a variety of sources can transport and introduce MP to surface water. Sewage solids from a variety of treatment processes have been analyzed directly [10,34,35]. Solids can serve as a sink for MP in wastewater treatment processes and

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**Fig. 1.** Heat map representing percent identity of total reported particles assigned a given polymer in different sources [9–12]. [MP are reported here are done so as the original authors did. Notably, authors reported Zn-sterate coated particles. Zn-sterate is a lubricant used for polymers and other materials. Cellulose-based polymers were also reported. Cellulose is a natural polymer but may be difficult to distinguish from regenerated cellulose (rayon)].
may be applied to land where MP can be transported to surface water in runoff. Consideration has also been given to land application of other materials to agricultural and in gardening soils including compost [36], and fertilizer [9]. Other sources of plastics exist in agricultural and home gardening (e.g., plastic mulch, landscape fabric) [37], but their impact on MP pollution has not been reported to the authors’ knowledge. Attempts to estimate the contribution of MP in sludge to soil (e.g., Ref. [10]) and amount of MP transported in runoff have been made (e.g., Ref. [37]). But, controlled runoff studies estimating the MP loading in agricultural runoff have not been published, to the authors’ knowledge.

Likewise, few data are available on the loading of MP in (urban) stormwater [38]. Stormwater may carry MP from tires and road markings [23,37,39], as well as other plastic litter. The potential for stormwater treatment systems to remove MP has not been reported, but removal of macroplastics has been studied (e.g., Ref. [40]).

Relatively fewer researchers have attempted to quantify atmospheric deposition of MP fibers [41], despite widespread anecdotal reports of careful measures being required to prevent contamination of field samples with fibers, likely from researchers clothing and/or air handling systems in laboratories. Comparison of up and
downstream fiber concentrations and fluxes surrounding Paris, FR indicated fibers did not increase downstream of the city [42]. Methods for polymer and natural fiber identification used in the textile industry have not been widely applied, including examination of fiber cross sections and longitudinal shapes, as well as flame tests that allow for visual and odor evaluation of the fibers on approach, within, and after removal from the flame, as well as for examination of the ashed residue [43–47]. To the authors’ knowledge, comparisons between the expected loads to freshwater from clothes washing versus shedding of fibers to the atmosphere have not been reported.

Several studies have linked MP abundance to urban land uses and population densities [22,48,49]. Xiong et al. [20] compared MP abundance in an urban lake in China to the abundance in the four rivers feeding into the lake to determine whether MP contamination in the lake originated from terrestrial sources or via surface water inputs. As MP abundances in the lake were higher than those in the rivers, it was concluded that terrestrial contamination, specifically improper solid waste disposal, was the primary source to the lake; sampling was performed during the wet season when transport from land would be expected in drawing this conclusion. Ballent et al. [49] explored MP in tributaries to a lake in Canada and mapped waste-water treatment facilities and plastic industries. Limited sample size prevented mass balance approaches for implicating sources and the authors noted “assigning particular origins to the microplastics is challenging due to their small size, fragmented nature, and the unknown range of possible sources.” Field observations have been used to indirectly indicate other sources than wastewater treatment plants in surface water including: the presence of MP in rivers upstream of wastewater treatment plants [31], in high concentrations in bed sediments without wastewater inputs [23], lack of mass balance in wastewater impacted surface waters [30], and comparisons of wastewater impacted versus unimpacted lakes [38]. Thus, mass balance approaches can be useful for indicating the magnitude of commonly targeted sources (i.e., wastewater treatment plants) and for highlighting the need to study nonpoint sources. Addressing these nonpoint sources may be more challenging given that they are more diffuse and diverse. Additionally, proper study design for mass balance approaches must be considered. Particularly, to accurately calculate loads, robust approaches with appropriate sampling techniques (likely composited samples) and sufficient sample sizes to capture variability are needed, which is challenging given analytical challenges that prevent high throughput sample analysis. In lieu of the ability to perform high resolution sampling, modeling the transport of MP [50] may also help fill in gaps in knowledge.

4. Can surface contaminants help to elucidate source?

While many MP analysis techniques use oxidation, enzymatic reactions, and/or acid washing to remove surface contaminants to allow for confirmation of the polymer composition, the contaminants present on the surface of MP particles may have potential utility in source tracking. One challenge with this potential approach is that surface contaminants adsorbed from the surrounding environment after MP release [51] should be considered in addition to the additives added during plastics manufacture to the polymer mixture which can be present on the surface (and within MP particles). Although challenging, identification of unique surface contaminants, including metals, organics and biofilms, could have utility for source tracking of MP pollution in freshwater systems if consistent relationships between surface contaminants and sources can be established and weathering processes understood. One significant challenge to consider is how to confirm that surface contaminants are adsorbed to MP rather than other particulate matter associated with MP particles.

Metals are commonly added as catalysts, pigments, and stabilizers during plastic manufacturing [52] therefore, targeting...
specific heavy metals may help indicate the source of MP pollution if these profiles are unique in different macroplastic sources. Wang et al. [53] summarized macroplastic heavy metal profiles then attempted to match MP and macroplastic heavy metal profiles. From their field sorption tests they concluded, that most heavy metals on the surface of the MPs they studied were from production rather than environmental sources. Thus, if the plastics produced with metals had unique applications there could be potential for linking the heavy metal profile to source, assuming environmental sorption and desorption processes did not interfere. There is evidence, however, from several studies demonstrating that virgin and aged MPs can adsorb metals during different stages of their transport pathway in the environment [18]. For example, metals extracted from MPs in the San Diego Bay area [54], identified by ICP-MS (inductively coupled plasma mass spectrometry), included toxic metals (Cd, Ni, Zn and Pb). The concentrations of these metals were more uniformly found on polymers than when MP particles were exposed to organic chemicals [54], which could vary by source. For sorbed heavy metals to have utility in source tracking the rates of weathering of these metals would need to be slower than transport and the concentrations present from production understood.

Different hydrophobic organic chemicals and persistent organic pollutants (POPs) are known to bind to MPs within the environment [55,56], especially in freshwater systems where the concentrations of these chemicals are expected to be higher due to proximity to the use of these chemicals [57]. The perceived risk of plastic to serve as a vector for these toxic chemicals to accumulate via the food chain has not been linked to plastic ingestion (as recently reviewed [58]). Identification of the POPs as well as polymer degradation products involve multi-step processes, including extraction of the organic(s) from the MP particles [59] followed by chromatography before chemical determination. It has been shown that hydrophobic organic compounds have greater affinity for MPs like polyethylene, polypropylene, and polyvinyl chloride than for natural sediments [51,57]. Changes to environmental conditions, such as residence time, temperature, pH and exposure to sunlight, will influence equilibrium dynamics between chemicals and MPs, impacting their accumulation and transport [12]. However, studying organic surface contaminants found on MPs may help lead to identification of the sources. Di and Wang [60] evaluated MP particles in surface waters and sediments in China. Organics or pharmaceutical intermediates were identified via Raman spectroscopy; the authors speculated that the origin of these organic substances may be due to the MP coming from hospitals or from sorption of pharmaceuticals to MP in the environment, given local discharges from hospitals.

A third surface contaminant that may have utility for MP source tracking is the growth of biofilms on MP surfaces. Given observations of unexpected microbial community members in MP biofilm in the ocean [61], there is some evidence that terrestrially sourced biofilms can be transported long distances. MP incubated in different water sources (i.e., river water versus wastewater) had unique biofilm microbial communities that clustered by source water rather than by polymer/morphology [62]. Similarly, marine biofilm community structures grown on PET the field varied by location and season [63]. More understanding of how robust biofilms are to weathering is needed to know if it is possible these biofilm community structures could help differentiate MP source.

5. Conclusions

Growing evidence suggests that MPs are present in freshwater systems and shoreline sediments worldwide and transfer of MPs from freshwater to marine systems is likely a significant source of marine microplastic contamination. Controlling the occurrence and transport of MPs in freshwater systems requires better understanding of MP sources and pathways including wastewater effluents, urban stormwater and agricultural runoff, atmospheric transport of fibers, and improper solid waste disposal adjacent to freshwater systems. MP characteristics, including morphology and polymer type may provide clues to improve our ability to source track microplastic contamination. In addition, surface contaminants including metals, organic compounds and biofilms may provide some evidence of microplastic source, but complications arise as environmental contaminants can also associate with freshwater MPs in situ. Challenges associated with source tracking based on these characteristics include limitations to accurate polymer identification, poor understanding of weathering processes, and limited understanding of microplastic transport mechanisms within freshwater systems.

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