

A Critical Review of Extraction and Identification Methods of Microplastics in Wastewater and Drinking Water

Dounia Elkhatib and Vinka Oyanedel-Craver*



Cite This: *Environ. Sci. Technol.* 2020, 54, 7037–7049



Read Online

ACCESS |



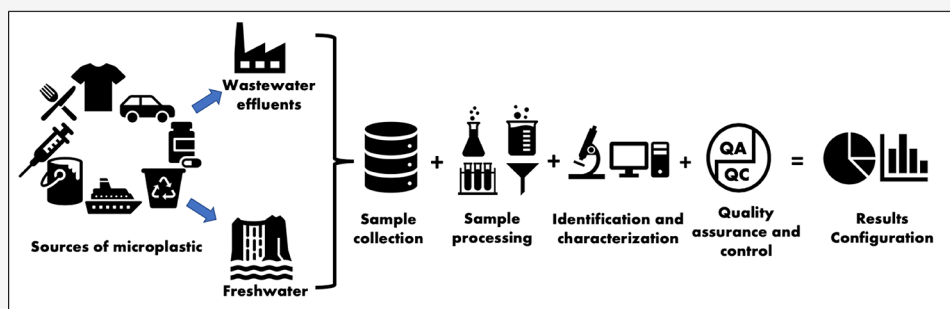
Metrics & More



Article Recommendations



Supporting Information



ABSTRACT: This critical review analyzes methodologies used to collect, quantify, and characterize microplastics in both wastewater and drinking water. Researchers used different techniques at all stages, from collection to characterization, for quantifying microplastics in urban water systems. This represents a barrier to comprehensively assess the current loads of microplastic and to compare the results obtained by such assessments. The compiled studies address microplastic contamination using four types of sample collection techniques, four methods for processing samples, and four techniques for characterizing microplastics. This results in significant discrepancies in each of the following: (1) reported concentrations in both wastewater effluents and drinking water, (2) microplastic characteristics (i.e., size, color, shape, and composition), and (3) quality control and assurance procedures. Finally, this review qualitatively evaluated the reports by the completeness of their data based on a ranking system using five criteria: sample collection, sample processing, quality control, identification technique, and results. The results of this ranking system clarify disparities between the studies.

1. INTRODUCTION

Microplastics (MPs) include acrylic, polyethylene, polypropylene, polyamide, and polyester fragments, among others, that are less than 5 mm in size.^{1–3}

Contamination of marine and freshwater environments by MPs has raised scientific and public concern due to the adverse impacts on aquatic life.^{1,4,5} The ingestion of MPs by aquatic organisms causes impaired reproduction, malnutrition, internal abrasions, and blockages.⁶ In addition, potentially toxic additives used to enhance the properties of plastics can be released into the water.⁷ Due to their hydrophobic surfaces, MPs can sorb persistent chemical pollutants (PCPs).^{8–10}

The presence of MPs in drinking water is a rising concern. Two identified sources of MPs in drinking water are the effluents from wastewater treatment plants (WWTPs) and urban and agricultural stormwater runoff.^{1,4,11–13} MPs have also been found in other products intended for human consumption,^{14–18} but information on adverse human health effects is only partially understood.^{15,19–21}

Previous studies have identified the presence of MPs in wastewater effluents and drinking water,^{22–25} but result comparisons are limited by the variability in the sampling

methods and identification techniques.^{24,26–29} Previous reviews have focused mainly on individual stages in the assessment of MPs,^{24,26–28} but an analysis of all of the stages (i.e., sample collection, processing, characterization, quality control and assurance, quantification) is required for a comprehensive understanding of the contamination caused by microplastics.

Presence of MPs has been frequently studied in marine ecosystems.^{3,5,24,27,30–36} However, there are limited data that involve wastewater and drinking water. Urban waters differ from the marine waters in matrix and organic contents, surface area, MPs sources, and sizes.

This critical review provides an analysis of all stages used in the assessment of MPs in both wastewater and drinking water, that is, sample collection, processing, identification, quality

Received: November 5, 2019

Revised: May 18, 2020

Accepted: May 20, 2020

Published: May 20, 2020



control and assurance, and results distribution. It also provides guidance for improving methodologies based on this analysis. The objectives are to (1) compare the methodologies used for the analysis of MPs in both wastewater and drinking water; (2) identify the research gaps and limitations of current techniques; and (3) develop a ranking system to evaluate the information provided by the current study and by future studies.

2. METHODOLOGY

The literature search used the following databases: ScienceDirect (<http://www.sciencedirect.com>), Scopus (<https://www.scopus.com>), Web of Science (<https://webofknowledge.com>), SpringerLink (<http://link.springer.com>), Wiley Online Library (<http://onlinelibrary.wiley.com>), Taylor & Francis Online (<http://www.tandfonline.com>), ACS Publications (<http://pubs.acs.org>), and RSC Publishing (<http://pubs.rsc.org>). The main keywords used in the search were *microplastic*, *wastewater*, *drinking water*, *freshwater*, *plastic waste*. This review paper cites 130 peer-reviewed publications or reports.

3. RESULTS

The first section describes the possible sources of MPs and their transport into the environment. The following two sections compile and analyze the procedures used during the assessment of MPs in wastewater and drinking water, from the collecting of samples to the reporting of results. Methods and procedures discussed in this review are limited to those reported in WWTPs and drinking water solely.

3.1. Sources and Fate of Microplastics. The global production of plastic exceeded 320 million tons in 2016,³⁷ with 9–21% of plastic waste either being recycled or incinerated; and the rest going to landfills or improperly disposed.³⁸ Improperly disposed plastic waste can result from unregulated anthropogenic waste, wind-blown waste from recycling facilities, or debris leaked during the collection and transportation of waste.³⁹

As a substantial constituent of plastic debris, MPs can be generated. Generally, MPs can have primary and secondary sources. Primary MPs are placed in cosmetics as abrasive or cleaning agents.^{40,41} They can also be generated by the breakdown of large plastic particles that break down when released in the environment.^{1,5} MPs are transported to the environment from both point or diffuse sources,⁴⁰ from which WWTPs are one of the main sources of MPs.^{25,42,43}

MPs released during household washing or industrial cleaning operations are transported through sewers to WWTPs.^{1,40,41,44,45} Due to their small sizes (<5 mm) and low density (<1.2 g/cm³),⁴⁶ only 50–98% of MPs are removed during primary treatment by the skimming and sedimentation processes,^{47,48} while an extra of 0.2–14% are removed during secondary treatment.^{49,50} A negligible amount of MPs are removed during tertiary treatment in some studies,^{49,51} whereas other studies showed an overall removal between 95 and 99%.^{48,52} Despite the high percentages removed, the total number of MPs released to natural bodies of water could be several million per day due to the large volumes of effluent discharged daily.^{23,48,53–55}

Many WWTPs receive a combined influent that consists of sewage and stormwater runoff.⁵⁶ In order to prevent hydraulic overload in the biological tank (secondary treatment) during extreme rain events, excess influent is either treated in a wet

weather protocol (mostly physicochemical treatment) or discharged without treatment in case of an overflow.⁵⁷ These discharges can carry large amounts of contaminants, including MPs.^{58–60}

Urban stormwater runoff contains MPs such as the wear of car tires and road markings that can be carried to the WWTPs runoff.^{61–65} An estimated 5–10% of the total amount of MPs in freshwater originate from tire wear.⁶³ Tire-wear is considered an important source of MPs with the particle sizes ranging from 5 μm to more than 100 μm ,^{63,66} and the majority of the particles ranging from 5 to 25 μm .^{63,66} Also, the paints used for road markings contain thermoplastic elastomers that can leach into the road runoff.⁶⁷ Similarly to other MPs, particles produced by the road markings and tire wear have low solubility and degradability, and they tend to accumulate in bodies of water.^{63–65,68}

Agricultural runoff from crops using biosolid-based fertilizers is a nonpoint source of MPs.^{59,65,69} MPs removed during the wastewater treatment process are retained in the sludge (biosolids), which can then be reused as farmland fertilizer.^{70–72} The sludge is treated before its agricultural use; however, MPs are not targeted for removal during treatment. Therefore, MPs can remain in the soils for decades after the application of sludge.^{73,74} MP particles retained in the sludge used for agriculture in North America has been reported to be between 4.4×10^4 and 3.0×10^5 tons per year.⁷⁵

Industrial products, such as drilling fluids, proppants, and cementing paste, can contain MPs.⁴⁵ Drilling fluids used in oil and gas exploration contain plastic microbeads, such as polytetrafluoroethylene (PTFE);⁷⁶ rust and paint removers utilize polyester scrubbers.⁷⁶ If these products are used in open space and not adequately contained, the MPs can end up in the aquatic environment.¹

Unregulated disposal of plastic products is a secondary source of microplastics.⁴⁵ Large plastic fragments get broken down through mechanical erosion, UV radiation, and biological degradation by bacteria or fungi.^{5,33} UV radiation degrades synthetic polymers of plastics in the environment by photo-oxidation,⁷⁷ with a degradation rate that depends on the composition of the polymer.⁷⁸ Physical degradation through abrasion, heating/cooling, freezing/thawing, and wetting/drying are other possible causes of the formation of MPs.⁷⁹ Ultimately, physical fragmentation can lead to the formation of micro- and nanoscale particles.⁸⁰ In addition, microorganisms, such as *Pseudomonas* sp. and *Bacillus brevis*, produce enzymes that are capable of promoting the hydrolysis of amides, esters, and urethane bonds, further degrading plastic particles.^{80–82}

Landfills are the most commonly used end-of-life disposal method for plastic waste in the United States (75.8%), followed by incineration at 15.8% and recycling at 8.4%.⁸³ Plastic waste in landfills may be exposed to extreme pH levels and temperatures, high salinity, microbial degradation, and physical stress.⁸⁴ These conditions lead to plastic fragmenting into MPs that can be leached from the landfill.^{85,86}

As described above, primary, and secondary sources of MPs contribute to the pollution of aquatic environments. Identifying the sources of MPs can be challenging since plastic particles travel long distances from their origin and undergo a series of transformations that change their physical and chemical characteristics.

3.2. Sampling and Identification of Microplastic in Wastewater. This section presents the compilation of studies of MPs in WWTPs conducted worldwide between 2011 and

Table 1. Comparison of MPs Sampling Techniques used in 22 Wastewater Studies

sampling method (% of studies using the method)	advantages	limitations
grab sample (40%) ^{53,55,89,90,147,48,51,91,100}	easy to perform, single time sample with less exposure to environmental contamination	samples may not be representative of the various characteristics of WW influents due to patterns of use and seasonal effects
extraction pump (20%) ^{49,54,55,92,95}	samples can provide a better representation of the varying properties of WW than grab samples; large volumes can be collected	sampling flow must be adjusted to the characteristics of the WW
composite sample (20%) ^{55,60,92-94}	the samples can represent the average performance of a WWTP during the collection period	time-consuming and requires multiple sampling trips, limited volumes can be collected
Neuston/plankton net (12%) ^{49,96,97}	easy to perform	smaller particles are underrepresented; possible cross-contamination due to the plastic net
custom method (8%) ^{48,52,99}	procedures developed in house by researchers	no consensus for use; results could be difficult to replicate

Table 2. Comparison of the Sample Processing Techniques used for MPs in Wastewater Studies

processing method (22 studies)	advantages	limitations
NOAA method (48%) ^{48,54,89,92-97,99,100}	organic matter is dissolved, resulting in clean MPs MPs used by several studies	might need more than one digestion step, increasing the time required different solutions were used to facilitate separation based on density through flotation; ZnCl ₂ and NaI had higher extraction efficiency than NaCl, but both are more expensive than NaCl
simple filtration (39%) ^{1,51-53,55,64,74,90,91}	easy, time-saving and low-cost	Difficulties in separating plastic particles from other organic or nonorganic particles.
centrifugation (9%) ^{49,89}	the method is easy and simple to use	fractioning and deformation of plastic particles, resulting in misrepresenting quantity, shape, and size
staining method (4%) ⁹⁹	easy to perform and low cost	false affirmation of some MP polymers; some algae and other organic detritus, such as seaweeds, wood, and feathers might be lightly stained giving false indications for MP

2019. These studies are categorized by location, sampling procedure, sample processing, identification and characterization techniques, and results (concentrations, quantities, and size distributions of the MPs). [Supporting Information Table S1](#) summarizes the information from all the studies discussed below.

3.2.1. Sampling Locations. Across multiple studies, ninety-three WWTPs were assessed for MP contamination. The results were summarized by countries and continents ([SI Table S3](#)). Most studies (94%) were performed in North America and Europe, however, this review only covered studies published in English-language journals. The abundance of MPs and their distribution in WWTPs varies by location with several environmental factors that affect their distribution and concentration. Factors include rainfall patterns, wind speed and direction, and other meteorological patterns.⁸⁷ Anthropogenic factors, such as population densities and daily human activities also affect the abundance and concentration of MPs. Wastewater streams in densely populated, urban areas are more likely to contain higher concentrations of MPs than wastewater streams in rural areas.¹ Poor waste management practices can also contribute to MP pollution since wastes placed in open dumps and uncontrolled landfills increase the production of MPs. Often, waste disposal in developing countries is poorly managed, leading to higher MP contamination in these areas.⁸⁸ The lack of studies related to MPs in developing countries can lead to underestimating their MP loads to the environment.

3.2.2. Collection and Processing Methods. **3.2.2.1. Sample Collection.** The studies included in this review identified four types of sample collection techniques, that is, grab samples, composite samples, extraction pumps, and Neuston nets. [Table 1](#) summarizes the studies presented in this section, including the advantages and limitations of each method.

Grab samples, which were used in 40% of the studies, are single samples taken at one time. The volumes of grab samples varied between 1 and 38 L, and containers included steel buckets,^{53,55} plastic containers,^{89,90} glass containers,^{1,51} and not noted in some studies.^{47,91}

Composite samples, which are collections of equal, discrete sample volumes taken at regular intervals over a period of time, were used in 20% of the studies.^{55,60,92-94} The time intervals between sample collections varied from 15 min to 2 h over a total period of 24 h. The volumes of composite samples were between 3.6 and 5 L,^{93,94} or was not reported.^{55,60,92}

Grab and composite samples were either vacuum-filtered onto membrane filters or passed through a stack of sieves ranging from 60 to 5000 μm .

The extraction pump technique was used in 20% of the studies.^{49,54,55,92,95} This sampling technique consists of using an electric pump to pump water from the wastewater stream directly onto a stack of sieves with mesh sizes ranging from 20 to 5000 μm . Flow rates varied from 2 to 22 L/min, and the sampling time varied from 2 to 24 h.^{49,54,55,92,95} The variability of both flow rates and sampling times led to sampling volumes ranging from 120 to 18 000 L. The differences in the flow rates are a function of the concentrations of solids in different streams and the mesh sizes of the sieves utilized. For example, the effluent from a WWTP has a low concentration of total solids, so it can be pumped at a high volumetric flow rate for long periods of time. However, the influent to a WWTP or an activated sludge tank has a higher concentration of total solids and lower flow rates are required.^{55,90} A few studies indicated that the flow rate varied during the collection period.^{49,54,55}

In 12% of the studies, MPs were collected using Neuston Nets or Plankton Nets.^{49,96,97} Neuston Nets consist of a net in a rectangular frame pulled by a rope which collects particles in the upper 10 cm of the water column. The mesh sizes of the

Table 3. Quality Assurance Procedures Applied during Sample Processing of MPs in 21 WWTPs

quality control measure	procedures applied during sample processing
blanks (54%) ^{48,52,54,55,60,74,90,92,93,95–97,100}	DI water was processed the same way as the samples as blanks
internal standards (21%) ^{89,91,92,97,99}	a known amount of MP polymer (e.g., polyethylene, polystyrene) was added as an internal standard
cleaning procedure (29%) ^{1,48,52,53,74,94,100}	all laboratory equipment and benches were cleaned; no synthetic clothing was worn during the experiment; no plastic tools were used; air circulation was avoided
control (33%) ^{48,52,53,74,89,93,94,100}	clean filter papers were placed in Petri dishes and exposed to the air in the laboratory during the processing time to account for atmospheric contamination

Table 4. Advantages and Limitations of the Methods Used for Identifying MPs in 23 WWTP Studies

techniques (% of studies)	advantages	limitations
Visual inspection/ microscope (50%) ^{47,49,51–55,60,74,90,92–97,99}	fast and easy identifies shape, size, and colors	lack of information on the plastic composition; not confirmative of plastic nature of the particle
FTIR (38%) ^{1,47,48,51–53,55,74,89,91–93,99}	identifies the composition of the polymer confirmation of the composition of the MP able to detect small plastic particles (~20 μm)	expensive tedious work and time-consuming to analyze all of the particles retained on the filter wavelength radiation can be a limiting detection factor
Raman (9%) ^{92,94,100}	identification of the composition of the polymer; confirmation of the composition of the MP; detection of small microplastics (1 μm) and nanoplastic (<1 μm)	expensive instrumentation time-consuming interference with pigments and contaminants
SEM/EDS (3%) ¹⁰³	clear and high-resolution images of particles facilitates differentiating between plastic and mineral particles due to the dominant inorganic elements (Si, Ca)	nonaffirmative results in plastic particles; lack of information on the type of polymer

nets used in the studies ranged between 150 and 330 μm ,^{49,96,97} limiting the detection of MPs to particles that are equal to or larger than the mesh size. The smaller sizes of MPs can be underestimated. Other concerns of Neuston Nets are atmospheric deposition and cross-contamination from the net itself.⁹⁸

In 8% of the studies, a custom sampling method was used that was developed in house (SI Table S1).^{48,55,99} Custom-sampling devices consist of a filtering device with an electric pump;⁵⁵ stacked units of mesh screens fixed to a base and placed inside a polyvinyl chloride (PVC) cover with the sampled water flowing through it;^{55,99} and a mobile pumping device made of a flexible PVC hose connected to a membrane pump, a polycarbonate filter housing containing a stainless steel cartridge filter and a flowmeter.⁴⁸

3.2.2.2. Sample Processing. After sampling, MPs are processed to separate them from other particles, such as organic and inorganic colloids. Table 2 presents a summary of the studies as well as the advantages and limitations of each of the processing techniques described below.

The National Ocean and Atmospheric Administration (NOAA) method¹⁰¹ is the most commonly used method for processing samples (48%).^{54,89,92,95–97,99} The procedure was designed for samples collected in marine environments; however, due to the complex matrix of wastewater and the presence of numerous particles of organic matter, this procedure was considered suitable by other nonmarine studies. This method involves digesting organic matter using hydrogen peroxide (H_2O_2) in the presence of an aqueous ferrous solution (Fe(II)) as a catalyst. The digestion step usually is followed by a separation step, which uses sodium chloride or zinc chloride solution to increase the density of the liquid

phase. This allows the low-density MPs to float, and the high-density particles to settle to the bottom. Then, the solution is filtered through mesh sizes varying from 0.7 to 125 μm (SI Table S1). A detailed description of the method is provided in the Supporting Information. The samples might require several digestion steps to dissolve all the attached organic matter; consequently, this process is time-consuming and more expensive than the other methods.

Thirty-nine percent of the studies used filtration method as the only treatment for the samples. Samples were either filtered directly through membrane filters or passed through sieves and followed by a vacuum filtration. The samples that passed through a stack of sieves in the range of 20–500 μm , were transferred using DI water into glass containers, and filtered using a vacuum pump.^{52,53,74,90,91} Membrane filters have pore sizes that vary between 0.1 and 11 μm .^{1,47,51–53,74,90,91} This method does not differentiate MPs from other organic particles.

Alternatively, some studies (10%) used centrifugation to process the samples, followed by sample filtration.^{49,89} Centrifugation conditions ranged from 4000 to 4500 rpm applied for a range of 2–20 min.^{49,89} The main drawback was that centrifugation may have deformed, compressed, or broken plastic particles, resulting in an inaccurate determination of the number of MPs.⁹²

Five percent of the studies used Rose Bengal staining solution.⁹⁹ The Rose Bengal solution stains organic particles, such as natural fibers and other colloidal organic particles, but not plastics, allowing a visual differentiation between plastic and nonplastic particles.⁹⁹ However, some studies have reported that some MP polymers (such as PVPP) were

stained by Rose bengal, while other natural particles such as starch and cellulose particles, were not stained.¹⁰²

Quality control and assurance procedures were inconsistent, with only 29% of the studies reported cleaning procedures for the equipment, glassware, and benches, among others.^{1,48,52,53,74,94,100} Table 3 shows that blanks, controls, and internal standards were used or reported by a limited number of studies. However, none of the studies used or reported quality control during field sampling to prevent cross-contamination; appropriate quality control would include wearing nonsynthetic coats, covering and preserving samples, careful sample shipping to control samples, and eliminating airborne contamination, among others. Some studies used plastic containers, nylon nets, coats made of synthetic fabrics, and used practices which can result in contamination of the samples.

3.2.3. Identification and Quantification Methods. After processing, particles retained on filters or sieves are counted and identified using several techniques. MPs can have various colors, shapes, sizes, and composition; therefore, multiple characterization techniques can be used. The most commonly reported identification methods were visual inspection using an optical microscope, Fourier-transform infrared spectroscopy (FTIR), Raman spectroscopy, and scanning electron microscopy (SEM). Table 4 summarizes the advantages and limitations of each one of the identification methods.

3.2.3.1. Visual Identification. Optical examination of MPs was used by 50% of the studies to identify plastic particles. Visual identification was done by the naked eye or by using an optical microscope with objectives ranging between 10× and 50×.^{54,60,90,95–97} In some cases, the microscope was coupled with image-analysis software, such as Histolab and Olympus stream.^{60,92} Although visual identification method is an easy and straightforward method, it is not a reliable identification of microplastics. This technique is susceptible to false positives resulting from interference by nonplastic particles; including cellulose, keratin, viscose rayon, coal/fly ash, and paint chips.^{4,104} Particles smaller than 100 μm and transparent particles are difficult to identify using this technique.

3.2.3.2. Raman Spectroscopy and Fourier-Transform Infrared Spectroscopy (FTIR). Raman spectroscopy (9% of studies^{92,94,100}) and FTIR (38%^{5,24,48,93,105–107}) were the most common analytical methods used to identify the composition of MPs, that is, polyethylene, polypropylene, polystyrene, or polyvinyl chloride.^{5,24,92,94,100,105–107} In both methods, molecules of the samples are investigated by vibrational spectroscopy producing a characteristic spectrum of the polymer that can be identified using a reference spectra library.^{26,108} For FTIR, the infrared radiation that irradiates the molecular structure of the particle is partially absorbed and measured in transmission or reflection mode.³ For Raman spectroscopy, a monochromatic laser source interacts with the molecules of the sample upon the scattering of light, resulting in characteristic molecular vibrations depending on the chemical structure of the component.^{109–111} Similar to the FTIR, Raman spectroscopy generates a spectrum that can be used to identify the polymer present in the particles.¹¹¹

The two modes of FTIR that can measure more than one fragment at a time are the attenuated total reflectance (ATR) mode and the focal plane array (FPA) mode.^{112,113} Larger particles (>500 μm) that can be handled with tweezers are usually analyzed using the ATR mode.²⁴ This type of analysis is not applicable for smaller particles. However, FPA-based,

micro-FTIR imaging can be used by combining FTIR with an optical microscope. FPA-FTIR can analyze particles collected on a filter by detecting each individually.¹¹⁴ However, micro-FTIR is a more expensive instrument and can be time-consuming when examining all of the MP particles on a filter.²⁴ Consequently, studies have analyzed small areas of the filters or confirmed the composition of a few MP particles.^{1,51–53,55,64,92,99} It is possible however to reduce the analysis time by creating an image of an entire filter in a single measurement using widefield imaging. Analysis times reported for this technique varied between 33 s/mm² to 111 min/mm².¹⁰⁹ The analysis times depend on the measurement settings including acquisition time, exposure time, number of accumulations, scan mapping points, and on the number of particles.¹⁰⁹ Another limitation of FTIR is that particles smaller than 20 μm cannot be detected.¹¹¹

An advantage of Raman spectroscopy is its ability to detect MP particles with sizes between 1 and 20 μm due to the smaller diameter of the laser beam as compared to the FTIR.^{5,24,106,107} Additionally, optical tweezers combined with Raman spectroscopy (i.e., Raman tweezers) demonstrated the potential of analyzing nanoplastics with sizes less than 1 μm.¹¹⁵ Raman spectroscopy has also the potential to analyze MPs from water samples.¹⁰⁹ However, fluorescence, due to pigments or additives in the MPs, interferes with the Raman spectra and may prevent the identification of the particles.¹¹¹ A limited number of studies (9%) used Raman spectroscopy for wastewater samples, which explains why the most commonly reported sizes of microplastic particles are 25 μm or larger⁹² (SI Table S1). The analysis time of Raman spectroscopy, which is in the range of 15–33 h/mm², is longer than that of FTIR.¹⁰⁹ Shorter measurement times (2 min/mm² to 55 min/mm²) were achieved using specific software (e.g., *Horiba, rapID*).¹⁰⁹

3.2.3.3. Scanning Electron Microscopy (SEM). A few studies (3%) used scanning electron microscopy (SEM), which visualizes the surface characteristics of the particles.¹⁰³ This technique provides high-resolution images of a sample by scanning the surface with a focused electron beam.¹¹⁶ The detailed sample images (>0.5 nm) allow differentiation between particles.¹¹⁷ However, SEM does not identify the composition of the polymer. In addition, the samples require special preparation, such as cleaning, drying, applying a conductive coating, and mounting the sample on a stub using conductive tape.¹⁰³

3.2.4. Reported Concentration and Properties of Microplastic. Units reported for MP are inconsistent in recording the sample location (influent, effluent, solids, upstream, downstream) and in the characteristics of the MPs (sizes, shapes, colors, and type of polymer). The various units that are used include MP particle number per volume (e.g., MP/L, MP/gal, MP/m³), particle number per mass (e.g., MP/kg), particle number per area (e.g., MP/m²), and concentration (e.g., mg/m³). While some studies provide shape and composition, others just reported the number of particles (SI Table S1). In addition, most studies performed an analysis of a subset of the MPs that were collected, implying that the total concentration can be determined by extrapolation. This leads to inaccurate results because it assumes that there was a consistent distribution of particles on the filter.

The following sections compare the results from the WWTP studies regarding MP concentrations, sizes, composition, and morphological characteristics.

3.2.4.1. Concentrations of Microplastic Particles. Due to units inconsistencies and the size distribution among the studies, the authors of this paper converted all MP concentrations into number of particles per volume (SI Table S4) when adequate information was provided.

Most of the studies (71%) reported MP sizes in the range of 100 to 125 μm ,^{47,49,51,53,54,60,90,92,95,97,99,103} but, in 25% of the studies, the sizes were 500 μm or larger.^{51,53,60,96,103} Using FTIR analysis, only 4% of the studies reported MP results for particles as small as 20 μm .⁴⁹ A few studies identified particles from 20 to 65 μm , but only reported the total number of particles irrespective of their sizes.^{51,55,90,91} Optical examination was the only analysis used for particles larger than 300 μm , while analyses of the compositions usually were included for particles smaller than 300 μm .

Most studies (63%) reported concentration values ranging from 8.0×10^{-1} to 5.0×10^2 MP/ m^3 . Twenty seven percent of the studies reported concentrations ranging from 1.0×10^3 to 6.0×10^3 MP/ m^3 in the wastewater effluents. A few studies (10%) reported high concentrations, ranging from 3.5×10^4 to 8.1×10^4 MP/ m^3 . As previously discussed, this variability was attributed to using different methods for sampling and identification. The levels of treatment (primary, secondary, and tertiary) at WWTPs can also affect concentrations. In general, higher MP removal rates are achieved at advanced treatment levels,^{48,52} with some exceptions.⁴⁹ Also, some advanced wastewater treatment processes can produce high MP removal rates including the membrane bioreactor (MBR) which removed 99.9% of MPs (reduced from 6.9×10^3 MP/ m^3 to 5.0×10^1 MP/ m^3),⁵² the rapid sand filter which removed 97% of the MPs (reduced from 7.0×10^2 to 2.0×10^1 MP/ m^3), and the dissolved air flotation process which removed 95% of the MPs (reduced from 2.0×10^3 to 1.0×10^2 MP/ m^3).⁵²

Only 39% of the studies reported the concentration of MP particles in the influent to the WWTP. Those concentrations, expressed as the number of particles per m^3 , mainly varied between 1.5×10^4 and 9.10×10^5 MPs/ m^3 .^{51,53,55,60,64,90} This 60-fold difference may have been due to the different geographical locations of the WWTPs, the volume and composition of the influent, the methods used to process and identify samples, and the presence of a combined sewage system.

3.2.4.2. Composition of MP Particles. The polymer composition of the MP particles was only reported when spectroscopy was used. However, a small number of the total extracted samples was analyzed.^{1,47,51–53,55,92,99} Therefore, percentages of the MP compositions were mostly extrapolations. Additionally, spectroscopy was used to analyze a fraction (6–18%) of the total area of the filter, whereas only a few particles were analyzed in the rest of studies.^{1,47,51–53,55,92,99} Table 6 presents the percentages of particles for the polymers that were identified most often, that is, polyester (PES), polyethylene (PE), polyethylene terephthalate (PET); nylon (PA), polypropylene (PP); and polystyrene (PS).

PES was the most reported polymer in WW effluents. The most common source of PES is the washing of synthetic clothes.¹¹⁸ The compositions of microplastics in personal care products and cleaning agents consist mostly of PE, PP, and PS particles that have an average sizes in the range of 150 to 330 μm .¹¹⁹ This matches the results found in terms of size (SI Table S4) and polymer distribution (Table 5).

Table 5. Percentage of the Number of Particles by Polymer^a

	PES	PET	PE	PA	acrylic	PP	alkyd	PS
average (%)	41.6	27.1	26.3	21.3	12	9.8	8	5.3
range (%)	11–67	4–51	4–70	16–28	7–17	3–16	8	4–10

^aPES = polyester, PET = polyethylene terephthalate, PE = polyethylene, PA = polyamide, PP = polypropylene, PS = polystyrene.^{1,47,52,53,55,99}

3.2.4.3. Morphology and Physical Characteristics. MP particles have been classified into categories according to their morphology as follows:^{3,120} plastic fragments or flakes (round, subround or angular), pellets (sphere, cylindrical, disks), filaments, plastic films, foamed plastic, beads, and Styrofoam. Colors reported included clear or transparent, white, white-cream, red, orange, blue, black, and others. These characteristics usually are assessed by using optical microscopy or scanning electron microscopy (SEM).²⁷ Eight studies reported the morphology of particles in the wastewater effluent.^{47,51,53,54,74,90,95,96} Table 6 shows the percent of particles

Table 6. Percentage of Morphology Distribution in Terms of Numbers of Mp Particles As Identified in Studies^{47,51,53,54,74,90,95,96}

MP morphology	fibers	fragment/ flake	film	beads	foam	pellets
average (%)	51.3	30.3	4.7	1.6	1.4	1.7
range (%)	18–80	18–68	1–10	0–3	1–2	1–3

by shapes identified in the studies that were reviewed. Fibers are the most dominant shape of MPs, and they most likely originate from washing synthetic clothes. Fragmented particles or flakes of particles were the second most common shape. Film, beads, foam, and pellet particles were less than 10% of the total MPs reported.

3.3. Sampling and Identification of Microplastics in Drinking Water. MPs have been identified in freshwater bodies,^{4,11,59,62,107,121} but there is very little information about their presence in water produced for human consumption.³⁴ A recent report by the World Health Organization reviewed the occurrence of MP in drinking water, with focusing on bottled and tap water and assessed the potential health risks to human.¹²² A search returned six peer-reviewed studies, and one report that had not been peer-reviewed. Four studies assessed the presence of MPs in bottled water,^{22,123–125} and three focused on the occurrence of MPs in tap water and drinking water treatment plants (DWTPs).^{126–128} Despite the small number of studies, there were substantial differences in the concentrations of MPs in all identified categories of drinking water.

The following sections compile the results found in the sampling, processing, identification methods as well as the MP concentrations in the reports concerning water for human consumption. SI Table S2 summarizes the relevant findings in the drinking water studies.

3.3.1. Sample Collection Techniques. Bottled water purchased globally was analyzed for MP contamination.^{22,123–125} The compositions of the packages included plastic, glass, carton, and high-density polyethylene (HDPE). This diversity of materials affects the results that were found for the MPs.

Another study relied on samples of tap water collected by volunteers stationed around the world.¹²⁸ The sampling procedure consisted of running the tap water for 1 min prior to filling a 500 mL HDPE bottle to the point of overflowing. The bottles were filled and emptied twice before being filled a third time and capped.¹²⁸ This was done to rinse the bottle prior to final sample collection.

Two studies collected water from the influent and effluent of drinking water treatment plants (DWTP) in the northwestern part of Germany¹²⁷ and in urban areas of the Czech Republic.¹²⁶ In addition, in one of the studies, a single household in the distribution water system of each DWTP was chosen for sampling at the meter and at the tap.¹²⁷ Volumes of drinking water samples (Table 7) were collected in glass containers and then filtered through 3 μm stainless steel cartridge filters¹²⁷ or 5 μm PTFE membrane filters.¹²⁶

Table 7 summarizes the volumes of the samples used by all studies.

3.3.2. Sample Processing. Bottled water samples were processed differently than wastewater samples since chemical digestion, density separation or centrifugation is not required. However, staining methods were used in two studies;^{125,128} while another study added ethylene diamine tetraacetic acid tetrasodium (EDTA) salt to reduce the number of precipitated minerals and facilitate the identification of plastic particles.¹²⁴ It should be noted that this method was not used or replicated by any other study that assessed MPs; therefore, further corroboration is needed. Two studies did not use any sample processing.^{22,123}

All studies filtered the samples under vacuum and stored the filters in sealed Petri dishes for further analysis. The Petri dishes were glass,^{124,126,127} polystyrol,²² or material not reported.^{123,128,129} Among the studies, the type of membrane filters used varied between cellulose nitrate,^{123,128} gold- or aluminum-coated polycarbonate,^{22,124} and glass fibers.¹²⁵ The pore size of the filters varied from 0.4 to 2.5 μm .

Samples collected from the DWTPs were digested using a H_2O_2 solution to remove any possible organic film.^{126,127} Then, the samples were filtered using 0.2 μm aluminum oxide filters.

Table 7 summarizes processing techniques that were used by the studies.

All studies used a laminar flow box to prevent any airborne contamination, and used a cleaning procedure for glassware and cotton lab coats to prevent any further external contamination. Blanks and controls were used in all studies while the samples were being processed in the lab.

3.3.3. Identification and Quantification Methods. The methods used to identify MP particles in drinking water also varied among the studies. Some used Raman microscopy ($N = 4$), some used FTIR ($N = 3$), and one used an optical microscope ($N = 1$). One study used both Raman microscopy and FTIR to analyze the samples.¹²⁶ Raman microscopy was carried out on the filters without prior processing of the samples,^{22,123} and for the particles between 1 and 10 μm after the sample had been digested.¹²⁶ After processing with digestion or staining, FTIR was used to confirm the composition of the polymer in the samples for particles larger than 10 μm .^{125–127} One study examined the stained filter under a dissecting microscope.¹²⁸

3.3.4. Concentrations and Composition of MP Particles. All studies reported the concentrations of MPs as either particles per liter or particles per cubic meter. The

Table 7. Summary of Processing Techniques Used in Assessing MPs in Drinking Water ($N = \text{Number of Studies}$) Analysis^{22,123–128}

type of water	sample volume	sample processing	identification technique	average concentration (MP/m ³)	comments
bottled water ($N = 4$)	0.5–2 L	vacuum filtration of samples ($N = 2$) ^{22,123} addition of Nile red solution ($N = 1$) ¹²⁵ addition of EDTA solution ($N = 1$) ¹²⁴	micro Raman spectroscopy identification by optical microscope - FTIR confirmation for some particles larger than 100 μm identification by micro-Raman spectroscopy	1.4×10^4 3.25×10^5 2.65×10^6	polymer composition detection - confirmation on MPs particles - detection of very small plastic particles (1 μm) FTIR was only used to provide more information on some specific polymers a small percentage (4.4%) of the sample area was analyzed
tap water ($N = 1$)	0.46–0.6 L	addition of rose-bengal solution ($N = 1$) ¹²⁸	identification by optical microscope for stained particles agitation by spatula for nonstained particles	4.5×10^3	false-positive results might be obtained
DWTP water ($N = 2$)	27 L 2,500 L	digestion using wet peroxide solution ($N = 2$) ^{126,127}	identification by FTIR microscope ¹²⁷ identification of particles larger than 10 μm by FTIR; and analysis of particles (1–10 μm) by Raman microscope ¹²⁶	4.0×10^{-1} 4.69×10^5	the whole filter was analyzed only 2.5% of the particles were analyzed per filter

concentrations (MPs/m³) varied significantly among the studies, that is, between 0 to 5.51×10^6 MPs/m³ for the single-use bottled water,^{22,123,124,129} 0 to 6.1×10^4 MPs/m³ for tap water,¹²⁸ 0 to 6.56×10^5 MPs/m³ for DWTP influent;¹²⁷ and 0 to 6.43×10^5 MPs/m³ for DWTP effluent.^{126,127} In general, single-use bottled water had higher MP concentrations than tap water or DWTP water, and, in some cases, the difference was orders of magnitude. Since the number of studies was limited and the reported range of concentrations varied significantly within the same study, no comparisons were possible. This variability was due to the heterogeneity of the samples that were tested, even within the same study.^{125,128}

SI Table S5 shows the MP concentrations (number of particles per cubic meter) reported in the studies by particle size.

SI Table S5 shows that the average concentrations for particles with sizes less than 100 μm (0 to 5.51×10^6 MP/m³) was larger than the concentration of particles greater than 100 μm (0 to 2.2×10^5 MP/m³). This was expected since drinking water undergoes several treatments and filtration processes that remove large MP particles.

The most common compositions of the MPs that were used for bottles and caps were PET (identified by four studies^{22,123,124,126}), PE (identified by three studies^{22,123,124}), and PES (identified by three studies^{22,125,127}). PES and PVC, standard materials in treatment plants and distribution systems, were detected in the tap water and DWTP water.¹²⁷

4. EVALUATION OF CURRENT STUDIES

Based on this review, we developed a ranking framework to determine whether the information provided in the studies could be compared. A quality assessment method was used to evaluate recent MP ingestion studies in aquatic biota using 10 criteria.¹³⁰ In this review, we adopted a simplified ranking framework to assess the completeness of MP studies in WWTPs. A highly ranked article would provide information in five categories: sample collection, sample processing, quality control (QA/QC), identification technique, and reports of the results of studies, that is, MP concentrations and compositions. The scores for each category ranged from 1 to 3, with 3 indicating the information is present, 2 indicating partially reported, and 1 indicating information is not present. The scores assigned for each study are summarized in SI Table S6. A more detailed overview of the scoring criteria is provided in SI Table S7.

Total scores for the studies ranged from 6 to 13. For a study to pass, it needed a score of 12 or above, meaning four out of five categories received a perfect rating. Only 24% of the total studies had passing scores. Those studies can be used for comparisons and for assessment of the MP contamination of wastewater. Note that quality control and identification methods were the two categories that were mostly under-reported.

The general trend of low scores in the studies casts doubt on the actual results that were found due to either under-estimating or overestimating the real concentrations of MPs.

The higher-scoring studies are the most recently published ones (2017–2019), suggesting an improvement in MP research methods. Three most recent studies provided complete information in all five categories.^{131–133} This ranking system can motivate researchers to use the best methods in order to eliminate some of the deficits associated with past and current studies.

5. DISCUSSION

Our results showed a lack of consistency among the studies with regards to quality assurance procedures, sample collection, sample processing, characterization and identification techniques, and reported results of MPs. Standardized procedures for all of the steps in the assessment of MPs in wastewater will increase the accuracy of the results, reduce time and effort required, and help to perform meaningful interpretations and comparisons between studies. Here, we discuss some recommendations for future studies.

Quality assurance and quality control methods are essential to prevent the contamination of the samples and ensure reliable results.¹³⁴ These methods include using a moist filter exposed to the air during the processing of the samples to account for airborne contamination,¹³⁵ a blank sample using DI water,^{54,96} spiking a sample with a known polymer,^{92,97} and using triplicate samples. Several other measures for control should be used during the processing of the samples, for example, the choice of plastic-free laboratory tools, the use of cotton lab coats, processing samples in a laminar flow chamber, and cleaning precautions for tools and the lab bench.^{135,136}

Choosing the appropriate technique for collecting samples, including the volumes of the samples, is challenging since each method has advantages and disadvantages. However, among the existing collection techniques, collecting with a grab sample seems to be the most appropriate. Samples should be collected in glass containers and sealed immediately on site, minimizing airborne contamination during sampling since the container will be underwater when the samples are taken.¹⁰ Then, wastewater samples can be digested and sorted through a stack of sieves with graduated mesh sizes. To prevent contamination, this process should be done in a laminar flow chamber in the lab.^{49,95} It is recommended that the mesh sizes of sieves be between 20 and 1000 μm , since sieves with smaller mesh sizes will be clogged by the contents of the wastewater.^{49,54} Then, filtrate water can be filtered through 10 and 1 μm membrane filters to retain the smaller particles. The filtrate water should be digested prior to filtration, as described later in the recommended processing method.

The removal efficiency of MPs at each treatment stages and their retention in sludge are essential to quantify the mass balances of MPs. This information helps to understand the fate of MPs in WWTPs. However, sampling from different units on the same day can be challenging due to the time required, the variable retention times of each unit, weather conditions, and the hours during which the facility is operated. Therefore, the grab-sample method seems to be the most convenient method.

The authors of this study identified four different techniques regarding sample processing methods. Since the wastewater matrix is very complicated, the digestion method developed by NOAA was appropriate to remove organic films.¹⁰¹ This method also has been frequently tested in water samples.^{89,101,137,138} The mixture of H₂O₂ and Fe²⁺ has been proven to purify wastewater due to its ability to remove organic compounds through oxidation.⁸⁹ The separation of MPs from other inorganic matter by density is best achieved using ZnCl₂.^{46,139} Filters can be chosen depending on the technique used to identify the MPs. For Raman spectroscopy analysis, the filter that has the best performance is aluminum polycarbonate, which helps reduce fluorescence and enhance the contrast between the MPs and the filter.¹⁴⁰ For micro-FTIR, the filter did not seem to interfere with the analysis,¹⁴¹ so membranes

such as nitrate cellulose, glass, and metal coated polycarbonate membranes can be used. The latter is also valid for any other identification technique. Since the current identification methods only detect particles that are larger than 1 μm , the sizes of the pores in the filters should be less than or equal to 1 μm . The filters should be stored in sealed glass Petri dishes to prevent any further contamination.

One of the main challenges in the analysis of MPs is identification. Visual examination of MPs using an optical microscope or SEM alone is not reliable. Visual inspection can be improved by using an automatic prescreening approach, named holographic plastic identification, that can distinguish MP accurately in heterogeneous pretreated water samples.¹⁴² However, subsequent determination of the composition of the polymer is required to confirm the identity of MP particles. Therefore, spectroscopy techniques, such as FTIR and Raman, are recommended for the identification of polymers. Raman microscopy has the advantage of detecting very small particles sizes (1 μm or less), whereas standard FTIR equipment can only detect particles larger than 20 μm . More recent techniques using a commercial setup that combines FTIR with atomic force microscopy AFM (AFM-IR) which allows the analysis of nanoplastic particles.¹⁴³ The time required to analyze the particles individually is one of the main limitations of these techniques,¹⁰⁹ but it can be reduced by using automated Raman mapping routines and particle detection software, enabling the rapid detection of MPs.¹⁰⁹

Significant variations in the concentrations and sizes of MPs have been reported in various studies, and we have identified the differences in the concentrations, sizes, units, morphologies, and compositions. However, comparing concentrations of MPs determined by various studies are meaningless when analyses are inconsistent. For example, filtering a sample through a 20 μm sieve will lead to a higher concentration of MPs than filtering the same sample through a 60 μm sieve. The use of large sieve sizes results in underestimating the total concentration of MPs. If all studies used the previous recommended techniques, compiling the results would be easy, and meaningful comparisons could be made among them. In addition, uniformity in units and terminologies is recommended, including expressing the concentration as the number of MPs/volume; expressing the total MPs in both the influent and effluent of WWTPs number of MPs/day; using a consistent range for the sizes of particles (μm); identifying the types of polymers, for example, PES, PE, PET, polyamide; and the classification of their shapes, for example, fiber, fragment, film, bead, foam, or pellet. Classification of MP based on morphology can help identifying their sources of origin. Fibers most likely originate from washing synthetic clothes, while films may originate from plastic bags or wrappers, and foams may be attributed to food containers or packaging.¹⁴⁴

The results and recommendations of this critical review can contribute to the U.S. Environmental Protection Agency's (USEPA's) report, which has identified and prioritized the scientific information required to understand the risks associated with MPs in the United States.¹⁴⁵ These recommendations include methods for the assessment of MPs, the identification of the sources and fate of MPs, and the risks to both human and ecological health posed by MPs. The USEPA's report is helpful in establishing work priorities while recognizing the deficits and needs, but there is still a need to generate planning scenarios regarding the unmet challenges associated with MPs. The recommendations put forth in this

critical review include some potential solutions to these challenges.

In studies of drinking water, the discrepancies also have been extensive, especially considering the limited number of studies. However, all studies of drinking water have avoided potential external sources of contamination by using blanks, controls, cleaning procedures, appropriate lab wear, and by performing experiments in a laminar flow box. Also, all but one of the studies have used spectroscopy methods to analyze drinking water samples, thereby attaining more accurate results.

The concentrations of MPs in drinking water, which range from 0 to 5505 MPs/L, were higher than their concentrations in wastewater, which range from 0.008 to 81 MPs/L. This difference is due mainly to the sizes of the MPs that were analyzed. Most wastewater studies analyzed particles larger than 100 μm , whereas drinking water studies analyzed particles between 1 and 500 μm . If studies conducted in WWTPs included the smaller-sized particles, their concentrations would have been higher. The processing of bottled water is another source of plastic contamination that can increase the concentrations of MPs in bottled drinking water. Regarding the effluents of DWTPs, there was a vast difference (4 orders of magnitude) in the concentrations between the two studies even though both studies used the same procedure for processing samples and the same spectroscopy techniques to identify the MPs. The main difference between the procedures was the volume of the samples. Normalized MPs concentrations were higher in the study with the larger volume, that is, 2500 L vs 27 L.

The volumes of drinking water samples used for the detection of MPs were generally very low, that is, 0.5–2 L, although there were two studies that used larger volumes.^{126,127} The concentrations of MPs in the small volumes were not representative of the actual concentrations of MPs in the water since the range of normalized MPs concentrations was extensive (0–5505 MPs/L). The concentrations reported were wide-ranging in the various studies due to the difference in sampling volumes, analyzing particles less than 10 μm in size, and analyzing only a fraction of the particles.

This review's recommendations for enhancing the methods for assessing MPs in both wastewater and drinking water are aimed at reducing the discrepancies among studies, creating a framework for legitimate comparisons of reported results, encouraging the performance of additional studies, and providing a better understanding of the issue of contamination caused by MPs.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.est.9b06672>.

- (1) Summary of the methodologies used in assessing MPs in WWTPs;
- (2) Summary of the methodologies used in assessing MPs in drinking water;
- (3) Number of WWTPs assessed for MP contamination in multiple studies and summarized by countries and continents;
- (4) MP concentrations expressed as the number of MP particles per volume in the final effluent for different particles sizes;
- (5) MP concentrations (number of particles per cubic meter) reported in the studies in consideration of the particles size;
- (6) Scores assigned for each study in five categories: sample collection,

sample processing, quality control (QA/QC), identification technique, and reporting studies results (MP concentrations and composition) (PDF)

AUTHOR INFORMATION

Corresponding Author

Vinka Oyanedel-Craver – Department of Civil and Environmental Engineering, University of Rhode Island, Kingston, Rhode Island, United States; Email: craver@uri.edu

Author

Dounia Elkhatib – Department of Civil and Environmental Engineering, University of Rhode Island, Kingston, Rhode Island, United States; orcid.org/0000-0002-2101-0238

Complete contact information is available at:

<https://pubs.acs.org/10.1021/acs.est.9b06672>

Author Contributions

The manuscript was written through the contributions of all authors. All authors have given approval to the final version of the manuscript.

Notes

The authors declare no competing financial interest.

REFERENCES

- (1) Browne, M.; Crump, P.; Niven, S.; Teuten, E.; Tonkin, A.; Galloway, T.; Thompson, R. Accumulation of Microplastic on Shorelines Worldwide: Sources and Sinks. *Environ. Sci. Technol.* **2011**, *45* (21), 9175.
- (2) Arthur, C.; Baker, J.; Bamford, H. *Proceedings of the International Research Workshop on the Occurrence, Effects and Fate of Microplastic Marine Debris*; NOAA Marine Debris Program, National Oceanic and Atmospheric Administration: U.S. Department of Commerce: Silver Spring, MD, 2009.
- (3) Hidalgo-Ruz, V.; Gutow, L.; Thompson, R. C.; Thiel, M. Microplastics in the marine environment: a review of the methods used for identification and quantification. *Environ. Sci. Technol.* **2012**, *46* (6), 3060.
- (4) Eriksen, M.; Mason, S.; Wilson, S.; Box, C.; Zellers, A.; Edwards, W.; Farley, H.; Amato, S. Microplastic pollution in the surface waters of the Laurentian Great Lakes. *Mar. Pollut. Bull.* **2013**, *77* (1–2), 177–182.
- (5) Cole, M.; Lindeque, P.; Halsband, C.; Galloway, T. S. Microplastics as contaminants in the marine environment: A review. *Mar. Pollut. Bull.* **2011**, *62* (12), 2588–2597.
- (6) Wright, S. L.; Kelly, F. J., Plastic and human health: a micro issue? *Environ. Sci. Technol.* **2017**, *51* (12).6634
- (7) Talsness, C. E.; Andrade, A. J. M.; Kuriyama, S. N.; Taylor, J. A.; Vom Saal, F. S. Components of plastic: experimental studies in animals and relevance for human health. *Philos. Trans. R. Soc., B* **2009**, *364* (1526), 2079.
- (8) Bakir, A.; Rowland, S. J.; Thompson, R. C. Enhanced desorption of persistent organic pollutants from microplastics under simulated physiological conditions. *Environ. Pollut.* **2014**, *185*, 16–23.
- (9) Mato, Y.; Isobe, T.; Takada, H.; Kanehiro, H.; Ohtake, C.; Kaminuma, T. Plastic resin pellets as a transport medium for toxic chemicals in the marine environment. *Environ. Sci. Technol.* **2001**, *35* (2), 318.
- (10) Wesch, C.; Bredimus, K.; Paulus, M.; Klein, R. Towards the suitable monitoring of ingestion of microplastics by marine biota: A review. *Environ. Pollut.* **2016**, *218*, 1200–1208.
- (11) Wagner, M.; Scherer, C.; Alvarez-Munoz, D.; Brennholt, N.; Bourrain, X.; Buchinger, S.; Fries, E.; Grosbois, C.; Klasmeier, J.; Marti, T.; Rodriguez-Mozaz, S.; Urbatzka, R.; Vethaak, A. D.; Winther-Nielsen, M.; Reifferscheid, G., Microplastics in freshwater ecosystems: what we know and what we need to know. *Environ. Sci. Eur.* **2014**, *26* (12). DOI: [10.1186/s12302-014-0012-7](https://doi.org/10.1186/s12302-014-0012-7)
- (12) Lasee, S.; Mauricio, J.; Thompson, W. A.; Karnjanapiboonwong, A.; Kasumba, J.; Subbiah, S.; Morse, A. N.; Anderson, T. A. Microplastics in a freshwater environment receiving treated wastewater effluent. *Integr. Environ. Assess. Manage.* **2017**, *13* (3), 528–532.
- (13) Eerkes-Medrano, D.; Thompson, R. C.; Aldridge, D. C. Microplastics in freshwater systems: A review of the emerging threats, identification of knowledge gaps and prioritisation of research needs. *Water Res.* **2015**, *75*, 63–82.
- (14) Yang, D.; Shi, H.; Li, L.; Li, J.; Jabeen, K.; Kolandhasamy, P. Microplastic Pollution in Table Salts from China. *Environ. Sci. Technol.* **2015**, *49* (22), 13622.
- (15) Smith, M.; Love, D.; Rochman, C.; Neff, R. Microplastics in Seafood and the Implications for Human Health. *Current Environmental Health Reports* **2018**, *5* (3), 375–386.
- (16) Liebezeit, G.; Liebezeit, E., Synthetic particles as contaminants in German beers. *Food Addit. Contam., Part A* **2014**, *31* (9).1574
- (17) Bouwmeester, H.; Hollman, P. C. H.; Peters, R. J. B. Potential Health Impact of Environmentally Released Micro- and Nanoplastics in the Human Food Production Chain: Experiences from Nanotoxicology. *Environ. Sci. Technol.* **2015**, *49* (15), 8932.
- (18) Rochman, C. M.; Tahir, A.; Williams, S. L.; Baxa, D. V.; Lam, R.; Miller, J. T.; Teh, F.-C.; Werorilangi, S.; Teh, S. J. Anthropogenic debris in seafood: Plastic debris and fibers from textiles in fish and bivalves sold for human consumption. *Sci. Rep.* **2015**, *5* (1), 14340.
- (19) Wright, S. L.; Kelly, F. J., Plastic and human health: a micro issue? *Environ. Sci. Technol.* **2017**, *51* (12).6634
- (20) Rist, S.; Carney Almroth, B.; Hartmann, N. B.; Karlsson, T. M. A critical perspective on early communications concerning human health aspects of microplastics. *Sci. Total Environ.* **2018**, *626*, 720–726.
- (21) Karbalaee, S.; Hanachi, P.; Walker, T. R.; Cole, M. Occurrence, sources, human health impacts and mitigation of microplastic pollution. *Environ. Sci. Pollut. Res.* **2018**, *25* (36), 36046.
- (22) Schymanski, D.; Goldbeck, C.; Humpf, H.-U.; Fürst, P. Analysis of microplastics in water by micro-Raman spectroscopy: Release of plastic particles from different packaging into mineral water. *Water Res.* **2018**, *129*, 154–162.
- (23) Prata, J. C. Microplastics in wastewater: State of the knowledge on sources, fate and solutions. *Mar. Pollut. Bull.* **2018**, *129* (1), 262–265.
- (24) Ivleva, N. P.; Wiesheu, A. C.; Niessner, R., Microplastic in Aquatic Ecosystems. 2017; Vol. 56, pp 1720–1739.
- (25) Ngo, P. L.; Pramanik, B. K.; Shah, K.; Roychand, R., Pathway, classification and removal efficiency of microplastics in wastewater treatment plants. *Environ. Pollut.* **2019**, *255* (Pt 2).113326
- (26) Shim, W. J.; Hong, S. H.; Eo, S. E. Identification methods in microplastic analysis: a review. *Anal. Methods* **2017**, *9* (9), 1384–1391.
- (27) Rocha-Santos, T.; Duarte, A. C. A critical overview of the analytical approaches to the occurrence, the fate and the behavior of microplastics in the environment. *TrAC, Trends Anal. Chem.* **2015**, *65*, 47–53.
- (28) Renner, G.; Schmidt, T. C.; Schram, J. Analytical methodologies for monitoring micro(nano)plastics: Which are fit for purpose? *Current Opinion in Environmental Science & Health* **2018**, *1*, 55–61.
- (29) Müller, Y. K.; Wernicke, T.; Pittroff, M.; Witzig, C. S.; Storck, F. R.; Klinger, J.; Zumbülte, N., Microplastic analysis-are we measuring the same? Results on the first global comparative study for microplastic analysis in a water sample. *Anal. Bioanal. Chem.* **2020**.412555
- (30) Rios Mendoza, L. M.; Karapanagioti, H.; Álvarez, N. R. Micro(nanoplastics) in the marine environment: Current knowledge and gaps. *Current Opinion in Environmental Science & Health* **2018**, *1*, 47–51.

- (31) Oberbeckmann, S.; LöDer, M. G. J.; Labrenz, M. Marine microplastic-associated biofilms – a review. *Environmental Chemistry* **2015**, *12* (5), 551.
- (32) Koelmans, A. A.; Bakir, A.; Burton, G. A.; Janssen, C. R. Microplastic as a Vector for Chemicals in the Aquatic Environment: Critical Review and Model-Supported Reinterpretation of Empirical Studies. *Environ. Sci. Technol.* **2016**, *50* (7), 3315.
- (33) Filella, M. Questions of size and numbers in environmental research on microplastics: methodological and conceptual aspects. *Environmental Chemistry* **2015**, *12* (5), 527.
- (34) Anbumani, S.; Kakkar, P., Ecotoxicological effects of microplastics on biota: a review. *Environ. Sci. Pollut. Res.* **2018**, 2514373
- (35) Hanvey, J. S.; Lewis, P. J.; Lavers, J. L.; Crosbie, N. D.; Pozo, K.; Clarke, B. O. A review of analytical techniques for quantifying microplastics in sediments. *Anal. Methods* **2017**, *9* (9), 1369–1383.
- (36) Wright, S. L.; Rowe, D.; Thompson, R. C.; Galloway, T. S., Microplastic ingestion decreases energy reserves in marine worms. **2013**; Vol. 23, pp R1031–R1033.
- (37) *PlasticsEurope Plastics - the Facts 2016: an Analysis of European Plastics Production, Demand and Waste Data*; PlasticsEurope, 2016.
- (38) Ravenscroft, M. Tackling ocean plastic waste. *Chemical Week* **2018**, *180* (10), 25.
- (39) Gourmelon, G. *Global Plastic Production Rises, Recycling Lags: New Worldwatch Institute Analysis Explores Trends In Global Plastic Consumption and Recycling*; Worldwatch Institute, 2015.
- (40) Siegfried, M.; Koelmans, A. A.; Besseling, E.; Kroeze, C. *Export of Microplastics from Land to Sea. A Modelling Approach*; Water Research, 2017.
- (41) Fendall, L. S.; Sewell, M. A. Contributing to marine pollution by washing your face: Microplastics in facial cleansers. *Mar. Pollut. Bull.* **2009**, *58* (8), 1225–1228.
- (42) Xu, X.; Jian, Y.; Xue, Y.; Hou, Q.; Wang, L. Microplastics in the wastewater treatment plants (WWTPs): Occurrence and removal. *Chemosphere* **2019**, *235*, 1089–1096.
- (43) Lv, X.; Dong, Q.; Zuo, Z.; Liu, Y.; Huang, X.; Wu, W.-M. Microplastics in a municipal wastewater treatment plant: Fate, dynamic distribution, removal efficiencies, and control strategies. *J. Cleaner Prod.* **2019**, *225*, 579–586.
- (44) Hernandez, L. M.; Yousefi, N.; Tufenkji, N. Are There Nanoplastics in Your Personal Care Products? *Environ. Sci. Technol. Lett.* **2017**, *4* (7), 280–285.
- (45) Duis, K.; Coors, A. Microplastics in the aquatic and terrestrial environment: sources (with a specific focus on personal care products), fate and effects. *Environ. Sci. Eur.* **2016**, *28* (1), 1–25.
- (46) Quinn, B.; Murphy, F.; Ewins, C. Validation of density separation for the rapid recovery of microplastics from sediment. *Anal. Methods* **2017**, *9* (9), 1491–1498.
- (47) Magnusson, K.; Norén, F. *Screening of Microplastic Particles in and down-Stream a Wastewater Treatment Plant*; Environmental Research Institute: 2014.
- (48) Mintenig, S. M.; Int-Veen, I.; Löder, M. G. J.; Primpke, S.; Gerdt, G. Identification of microplastic in effluents of waste water treatment plants using focal plane array-based micro-Fourier-transform infrared imaging. *Water Res.* **2017**, *108*, 365–372.
- (49) Carr, S. A.; Liu, J.; Tesoro, A. G. Transport and fate of microplastic particles in wastewater treatment plants. *Water Res.* **2016**, *91*, 174–182.
- (50) Sun, J.; Dai, X.; Wang, Q.; van Loosdrecht, M. C. M.; Ni, B.-J. Microplastics in wastewater treatment plants: Detection, occurrence and removal. *Water Res.* **2019**, *152*, 21–37.
- (51) Leslie, H. A.; Brandsma, S. H.; van Velzen, M. J. M.; Vethaak, A. D., Microplastics en route: Field measurements in the Dutch river delta and Amsterdam canals, wastewater treatment plants, North Sea sediments and biota. *Environ. Int.* **2017**, 101133
- (52) Talvitie, J.; Mikola, A.; Koistinen, A.; Setälä, O. Solutions to microplastic pollution – Removal of microplastics from wastewater effluent with advanced wastewater treatment technologies. *Water Res.* **2017**, *123*, 401–407.
- (53) Murphy, F.; Ewins, C.; Carbonnier, F.; Quinn, B. Wastewater Treatment Works (WwTW) as a Source of Microplastics in the Aquatic Environment. *Environ. Sci. Technol.* **2016**, *50* (11), 5800.
- (54) Mason, S. A.; Garneau, D.; Sutton, R.; Chu, Y.; Ehmann, K.; Barnes, J.; Fink, P.; Papazissimos, D.; Rogers, D. L. Microplastic pollution is widely detected in US municipal wastewater treatment plant effluent. *Environ. Pollut.* **2016**, *218*, 1045–1054.
- (55) Talvitie, J.; Mikola, A.; Setälä, O.; Heinonen, M.; Koistinen, A. How well is microlitter purified from wastewater? – A detailed study on the stepwise removal of microlitter in a tertiary level wastewater treatment plant. *Water Res.* **2017**, *109*, 164–172.
- (56) United States Environmental Protection Agency. Polluted Runoff: Nonpoint Source Pollution. <https://www.epa.gov/nps/what-nonpoint-source>.
- (57) United States Environmental Protection Agency. Combined Sewer Overflows. <https://www3.epa.gov/region1/eco/uep/cso.html>.
- (58) Hayley, T. O.; Steven, R. C.; Deborah, K. D.; Katherine, M. H.; Melinda, J. B.; Sandra, L. M. High levels of sewage contamination released from urban areas after storm events: A quantitative survey with sewage specific bacterial indicators. *PLoS Medicine* **2018**, *15* (7), No. e1002614.
- (59) Free, C. M.; Jensen, O. P.; Mason, S. A.; Eriksen, M.; Williamson, N. J.; Boldgiv, B. High-levels of microplastic pollution in a large, remote, mountain lake. *Mar. Pollut. Bull.* **2014**, *85* (1), 156–163.
- (60) Dris, R.; Gasperi, J.; Rocher, V.; Saad, M.; Renault, N.; Tassin, B. Microplastic contamination in an urban area: a case study in Greater Paris. *Environmental Chemistry* **2015**, *12* (5), 592.
- (61) Pieter Jan, K.; Ansje, J. L.; Frank, G. A. J. V. B.; Ad, M. J. R. Wear and Tear of Tyres: A Stealthy Source of Microplastics in the Environment. *Int. J. Environ. Res. Public Health* **2017**, *14* (10), 1265.
- (62) Horton, A. A.; Walton, A.; Spurgeon, D. J.; Lahive, E.; Svendsen, C. Microplastics in freshwater and terrestrial environments: Evaluating the current understanding to identify the knowledge gaps and future research priorities. *Sci. Total Environ.* **2017**, *586*, 127–141.
- (63) Kole, P. J.; Löhr, A. J.; Van Belleghem, F. G. A. J.; Ragas, A. M. J., Wear and Tear of Tyres: A Stealthy Source of Microplastics in the Environment. *Int. J. Environ. Res. Public Health* **2017**, *14* (10), 1265.
- (64) Magnusson, K. N.; Fredrik, N. Screening of microplastic particles in and down-stream a wastewater treatment plant; Environmental Research Institute: 2014.
- (65) Lassen, C.; Hansen, S. F.; Magnusson, K.; Hartmann, N. B.; Rehne Jensen, P.; Nielsen, T. G.; Brinch, A. *Microplastics: Occurrence, Effects, And Sources of Releases to the Environment in Denmark*; Danish Environmental Protection Agency: Copenhagen 2015.
- (66) Kreider, M. L.; Panko, J. M.; McAttee, B. L.; Sweet, L. I.; Finley, B. L. Physical and chemical characterization of tire-related particles: Comparison of particles generated using different methodologies. *Sci. Total Environ.* **2010**, *408* (3), 652–659.
- (67) Vogelsang, C. L.; L., Amy; Sundvor, I.; Umar, M. *Microplastics in Road Dust - Characteristics, Pathways and Measures*; Norwegian Institute for Water Research, 2018.
- (68) OSPAR Commission. *Assessment Document of Land-Based Inputs of Microplastics in the Marine Environment*, 2017.
- (69) Anderson, J. C.; Park, B. J.; Palace, V. P. Microplastics in aquatic environments: Implications for Canadian ecosystems. *Environ. Pollut.* **2016**, *218*, 269–280.
- (70) Zhang, Q.; Hu, J.; Lee, D.-J.; Chang, Y.; Lee, Y.-J. Sludge treatment: Current research trends. *Bioresour. Technol.* **2017**, *243*, 1159–1172.
- (71) Kacprzak, M.; Neczaj, E.; Fijałkowski, K.; Grobelak, A.; Grosser, A.; Worwag, M.; Rorat, A.; Brattebo, H.; Almás, Á.; Singh, B. R. Sewage sludge disposal strategies for sustainable development. *Environ. Res.* **2017**, *156*, 39–46.
- (72) Bertanza, G.; Canato, M.; Laera, G. Towards energy self-sufficiency and integral material recovery in waste water treatment plants: Assessment of upgrading options. *J. Cleaner Prod.* **2018**, *170*, 1206–1218.

- (73) Zubris, K. A. V.; Richards, B. K. Synthetic fibers as an indicator of land application of sludge. *Environ. Pollut.* **2005**, *138* (2), 201–211.
- (74) Mahon, A. M.; O'Connell, B.; Healy, M. G.; O'Connor, I.; Officer, R.; Nash, R.; Morrison, L. Microplastics in Sewage Sludge: Effects of Treatment. *Environ. Sci. Technol.* **2017**, *51* (2), 810.
- (75) Nizzetto, L.; Futter, M.; Langaas, S. Are Agricultural Soils Dumps for Microplastics of Urban Origin? *Environ. Sci. Technol.* **2016**, *50* (20), 10777.
- (76) Sundt, P.; Schulze, P.-E.; Syversen, F. *Sources of Microplastics-Pollution to the Marine Environment*; Norwegian Environment Agency Miljødirektoratet: Norway, 2014.
- (77) Andrady, A. L. Microplastics in the marine environment. *Mar. Pollut. Bull.* **2011**, *62* (8), 1596–1605.
- (78) Klein, S.; D., Ian; Eubeler, Jan; Knepper, Thomas, Analysis, Occurrence, and Degradation of Microplastics in the Aqueous Environment. In *Freshwater Microplastics. The Handbook of Environmental Chemistry*; Springer: Cham, Switzerland, 2017; Vol. 58
- (79) Gewert, B.; Plassmann, M. M.; Macleod, M. Pathways for degradation of plastic polymers floating in the marine environment. *Environ. Sci.: Processes Impacts* **2015**, *17* (9), 1513–1521.
- (80) Klein, S.; Dimzon, I.; Eubeler, J.; Knepper, T. Analysis, Occurrence, and Degradation of Microplastics in the Aqueous Environment. In *Freshwater Microplastics. The Handbook of Environmental Chemistry*; Springer: Cham, Switzerland, 2017; Vol. 58.
- (81) Singh, B.; Sharma, N. Mechanistic implications of plastic degradation. *Polym. Degrad. Stab.* **2008**, *93* (3), 561–584.
- (82) Müller, R.-J.; Kleeberg, I.; Deckwer, W.-D. Biodegradation of polyesters containing aromatic constituents. *J. Biotechnol.* **2001**, *86* (2), 87–95.
- (83) National Overview: Facts and Figures on Materials, Wastes and Recycling. <https://www.epa.gov/facts-and-figures-about-materials-waste-and-recycling/national-overview-facts-and-figures-materials>.
- (84) Kjeldsen, P.; Barlaz, M. A.; Rooker, A. P.; Baun, A.; Ledin, A.; Christensen, T. H. Present and Long-Term Composition of MSW Landfill Leachate: A Review. *Crit. Rev. Environ. Sci. Technol.* **2002**, *32* (4), 297–336.
- (85) He, P.; Chen, L.; Shao, L.; Zhang, H.; Lü, F. Municipal solid waste (MSW) landfill: A source of microplastics? -Evidence of microplastics in landfill leachate. *Water Res.* **2019**, *159*, 38–45.
- (86) Sundt, P.; S., Per-Erik; Syversen, Frode *Sources of Microplastics-Pollution to the Marine Environment*; Norwegian Environment Agency Miljødirektoratet: Norway, 2014.
- (87) Shahul Hamid, F.; Bhatti, M. S.; Anuar, N.; Anuar, N.; Mohan, P.; Periathamby, A. Worldwide distribution and abundance of microplastic: How dire is the situation? *Waste Manage. Res.* **2018**, *36* (10), 873–897.
- (88) Our World in Data; Ritchie, H. R. M. *Plastic Pollution*; Our World in Data: 2018.
- (89) Tagg, A. S.; Harrison, J. P.; Ju-Nam, Y.; Sapp, M.; Bradley, E. L.; Sinclair, C. J.; Ojeda, J. J. Fenton's reagent for the rapid and efficient isolation of microplastics from wastewater. *Chem. Commun.* **2017**, *53* (2), 372–375.
- (90) Michielssen, M. R.; Michielssen, E. R.; Ni, J.; Duhaime, M. B. Fate of microplastics and other small anthropogenic litter (SAL) in wastewater treatment plants depends on unit processes employed. *Environ. Sci.: Water Res. Technol.* **2016**, *2* (6), 1064–1073.
- (91) Majewsky, M.; Bitter, H.; Eiche, E.; Horn, H. Determination of microplastic polyethylene (PE) and polypropylene (PP) in environmental samples using thermal analysis (TGA-DSC). *Sci. Total Environ.* **2016**, *568*, 507–511.
- (92) Dyachenko, A.; Mitchell, J.; Arsem, N. Extraction and identification of microplastic particles from secondary wastewater treatment plant (WWTP) effluent. *Anal. Methods* **2017**, *9* (9), 1412–1418.
- (93) Conley, K.; Clum, A.; Deepe, J.; Lane, H.; Beckingham, B. Wastewater treatment plants as a source of microplastics to an urban estuary: Removal efficiencies and loading per capita over one year. *Water research X* **2019**, *3*, 100030.
- (94) Gündoğdu, S.; Çevik, C.; Güzel, E.; Kilercioğlu, S. Microplastics in municipal wastewater treatment plants in Turkey: a comparison of the influent and secondary effluent concentrations. *Environ. Monit. Assess.* **2018**, *190* (11), 626.
- (95) Sutton, R.; Mason, S. A.; Stanek, S. K.; Willis-Norton, E.; Wren, I. F.; Box, C. Microplastic contamination in the San Francisco Bay, California. *Mar. Pollut. Bull.* **2016**, *109* (1), 230–235.
- (96) McCormick, A. R.; Hoellein, T. J.; London, M. G.; Hittie, J.; Scott, J. W.; Kelly, J. J. Microplastic in surface waters of urban rivers: concentration, sources, and associated bacterial assemblages. *Ecosphere* **2016**, *7* (11), n/a–n/a.
- (97) Estabhanati, S.; Fahrenfeld, N. L. Influence of wastewater treatment plant discharges on microplastic concentrations in surface water. *Chemosphere* **2016**, *162*, 277–284.
- (98) Barrows, A. P. W.; Neumann, C. A.; Berger, M. L.; Shaw, S. D. Grab vs. neuston tow net: a microplastic sampling performance comparison and possible advances in the field. *Anal. Methods* **2017**, *9* (9), 1446–1453.
- (99) Ziajahromi, S.; Neale, P. A.; Rintoul, L.; Leusch, F. D. L. Wastewater treatment plants as a pathway for microplastics: Development of a new approach to sample wastewater-based microplastics. *Water Res.* **2017**, *112*, 93–99.
- (100) Wolff, S.; Kerpen, J.; Prediger, J.; Barkmann, L.; Müller, L. Determination of the microplastics emission in the effluent of a municipal waste water treatment plant using Raman microspectroscopy. *Water research X* **2019**, *2*, 100014.
- (101) Masura, J.; Baker, J.; Foster, G.; Arthur, C. *Analysis of Microplastics in the Marine Environment: Recommendations for Quantifying Synthetic Particles in Waters and Sediments*; NOAA Technical Memorandum, 2015.
- (102) Lachenmeier, D. W.; K., Jelena; Noack, D.; Kuballa, Thomas Microplastic identification in German beer – an artefact of laboratory contamination? *Deutsche Lebensmittel-Rundschau.* **2015**.
- (103) Kalčíková, G.; Alič, B.; Skalar, T.; Bundschuh, M.; Gotvajn, A. Ž. Wastewater treatment plant effluents as source of cosmetic polyethylene microbeads to freshwater. *Chemosphere* **2017**, *188*, 25–31.
- (104) Lenz, R.; Enders, K.; Stedmon, C. A.; Mackenzie, D. M. A.; Nielsen, T. G. A critical assessment of visual identification of marine microplastic using Raman spectroscopy for analysis improvement. *Mar. Pollut. Bull.* **2015**, *100* (1), 82–91.
- (105) Murray, F.; Cowie, P. R. Plastic contamination in the decapod crustacean *Nephrops norvegicus* (Linnaeus, 1758). *Mar. Pollut. Bull.* **2011**, *62* (6), 1207–1217.
- (106) Van Cauwenberghe, L.; Janssen, C. R. Microplastics in bivalves cultured for human consumption. *Environ. Pollut.* **2014**, *193*, 65–70.
- (107) Imhof, H. K.; Laforsch, C.; Wiesheu, A. C.; Schmid, J.; Anger, P. M.; Niessner, R.; Ivleva, N. P. Pigments and plastic in limnetic ecosystems: A qualitative and quantitative study on microparticles of different size classes. *Water Res.* **2016**, *98*, 64–74.
- (108) Hummel, D. O. *Atlas of Plastics Additives: Analysis by Spectrometric Methods*; Springer-Verlag: Berlin, Heidelberg, New York.: University of Cologne, Germany, 2003; Vol. 125, pp 1441–1441.
- (109) Araujo, C. F.; Nolasco, M. M.; Ribeiro, A. M. P.; Ribeiro-Claro, P. J. A. Identification of microplastics using Raman spectroscopy: Latest developments and future prospects. *Water Res.* **2018**, *142*, 426–440.
- (110) Collard, F.; Gilbert, B.; Eppe, G.; Parmentier, E.; Das, K. Detection of Anthropogenic Particles in Fish Stomachs: An Isolation Method Adapted to Identification by Raman Spectroscopy. *Arch. Environ. Contam. Toxicol.* **2015**, *69* (3), 331–339.
- (111) Käßler, A.; Fischer, D.; Oberbeckmann, S.; Schernewski, G.; Labrenz, M.; Eichhorn, K.-J.; Voit, B. Analysis of environmental microplastics by vibrational microspectroscopy: FTIR, Raman or both? *Anal. Bioanal. Chem.* **2016**, *408* (29), 8377–8391.

- (112) Löder, M. G. J. G. *Gunnar Methodology Used for the Detection and Identification of Microplastics—A Critical Appraisal*; University of Bayreuth, Germany, 2015.
- (113) Primpke, S.; Lorenz, C.; Rascher-Friesenhausen, R.; Gerdts, G. An automated approach for microplastics analysis using focal plane array (FPA) FTIR microscopy and image analysis. *Anal. Methods* **2017**, *9* (9), 1499–1511.
- (114) Harrison, J. P.; Ojeda, J. J.; Romero-Gonzalez, M. E., The applicability of reflectance micro-Fourier-transform infrared spectroscopy for the detection of synthetic microplastics in marine sediments. *Sci. Total Environ.* **2011**.
- (115) Gillibert, R.; Balakrishnan, G.; Deshoules, Q.; Tardivel, M.; Magazzù, A.; Donato, M. G.; Maragò, O. M.; Lamy de La Chapelle, M.; Colas, F.; Lagarde, F.; Gucciardi, P. G. Raman Tweezers for Small Microplastics and Nanoplastics Identification in Seawater. *Environ. Sci. Technol.* **2019**, *53* (15), 9003.
- (116) Zarfl, C., Promising techniques and open challenges for microplastic identification and quantification in environmental matrices. *Anal. Bioanal. Chem.* **2019**, 4113743
- (117) Wang, W.; Wang, J. Investigation of microplastics in aquatic environments: An overview of the methods used, from field sampling to laboratory analysis. *TrAC, Trends Anal. Chem.* **2018**, *108*, 195–202.
- (118) Napper, I. E.; Thompson, R. C. Release of synthetic microplastic plastic fibres from domestic washing machines: Effects of fabric type and washing conditions. *Mar. Pollut. Bull.* **2016**, *112* (1–2), 39–45.
- (119) Napper, I. E.; Bakir, A.; Rowland, S. J.; Thompson, R. C. Characterisation, quantity and sorptive properties of microplastics extracted from cosmetics. *Mar. Pollut. Bull.* **2015**, *99* (1–2), 178–185.
- (120) *Guide to Microplastic Identification*; Marine and Environmental Research Institute: Blue Hill, ME.
- (121) Wang, W.; Ndungu, A. W.; Li, Z.; Wang, J. Microplastics pollution in inland freshwaters of China: A case study in urban surface waters of Wuhan, China. *Sci. Total Environ.* **2017**, *575*, 1369–1374.
- (122) *Microplastics in Drinking-Water*; World Health Organization: Geneva, 2019.
- (123) Wiesheu, A. C.; Anger, P. M.; Baumann, T.; Niessner, R.; Ivleva, N. P. Raman microspectroscopic analysis of fibers in beverages. *Anal. Methods* **2016**, *8* (28), 5722–5725.
- (124) Oßmann, B. E.; Sarau, G.; Holtmannspötter, H.; Pischetsrieder, M.; Christiansen, S. H.; Dicke, W. Small-sized microplastics and pigmented particles in bottled mineral water. *Water Res.* **2018**, *141*, 307–316.
- (125) Mason, S. A.; W., Victoria; Neratko, J. *Synthetic Polymer Contamination in Bottled Water*; State University of New York at Fredonia, Department of Geology & Environmental Sciences, 2018.
- (126) Pivokonsky, M.; Cermakova, L.; Novotna, K.; Peer, P.; Cajthaml, T.; Janda, V. Occurrence of microplastics in raw and treated drinking water. *Sci. Total Environ.* **2018**, *643*, 1644–1651.
- (127) Mintenig, S. M.; Löder, M. G. J.; Primpke, S.; Gerdts, G. Low numbers of microplastics detected in drinking water from ground water sources. *Sci. Total Environ.* **2019**, *648*, 631–635.
- (128) Kosuth, M.; Mason, S. A.; Wattenberg, E. V. Anthropogenic contamination of tap water, beer, and sea salt. *PLoS One* **2018**, *13* (4), No. e0194970.
- (129) Mason, S. A.; Welch, V.; Neratko, J. *Synthetic Polymer Contamination in Bottled Water*; State University of New York at Fredonia, Department of Geology & Environmental Sciences: 2018.
- (130) Hermsen, E.; Mintenig, S. M.; Besseling, E.; Koelmans, A. A. Quality Criteria for the Analysis of Microplastic in Biota Samples: A Critical Review. *Environ. Sci. Technol.* **2018**, *52* (18), 10230.
- (131) Magni, S.; Binelli, A.; Pittura, L.; Avio, C. G.; Della Torre, C.; Parenti, C. C.; Gorbi, S.; Regoli, F. The fate of microplastics in an Italian Wastewater Treatment Plant. *Sci. Total Environ.* **2019**, *652*, 602–610.
- (132) Edo, C.; González-Pleiter, M.; Leganés, F.; Fernández-Piñas, F.; Rosal, R., Fate of microplastics in wastewater treatment plants and their environmental dispersion with effluent and sludge. *Environ. Pollut.* **2020**, 259.113837
- (133) Blair, R. M.; Waldron, S.; Gauchotte-Lindsay, C., Average daily flow of microplastics through a tertiary wastewater treatment plant over a ten-month period. *Water Res.* **2019**, 163.114909
- (134) Galgani, F. H. G.; Werner, S.; Oosterbaan, L.; Nilsson, P.; Fleet, D.; Kinsey, S.; Thompson, R.; van Franeker, J.; Vlachogianni, T.; Scoullou, M.; Veiga, J.; Palatinus, A.; Matiddi, M.; Maes, T.; Korpinen, S.; Budziak, A.; Leslie, H.; Gago, J.; Liebezeit, G. *Guidance on Monitoring of Marine Litter in European Seas*; Institute for Environment and Sustainability, 2013.
- (135) Wesch, C.; Elert, A. M.; Wörner, M.; Braun, U.; Klein, R.; Paulus, M. Assuring quality in microplastic monitoring: About the value of clean-air devices as essentials for verified data. *Sci. Rep.* **2017**, *7* (1), 5424.
- (136) Underwood, A. J.; Chapman, M. G.; Browne, M. A. Some problems and practicalities in design and interpretation of samples of microplastic waste. *Anal. Methods* **2017**, *9* (9), 1332–1345.
- (137) Zhao, S.; Zhu, L.; Wang, T.; Li, D. Suspended microplastics in the surface water of the Yangtze Estuary System, China: First observations on occurrence, distribution. *Mar. Pollut. Bull.* **2014**, *86* (1–2), 562–568.
- (138) Liebezeit, G.; Dubaish, F. Microplastics in Beaches of the East Frisian Islands Spiekeroog and Kachelotplate. *Bull. Environ. Contam. Toxicol.* **2012**, *89* (1), 213–217.
- (139) Coppock, R. L.; Cole, M.; Lindeque, P. K.; Queirós, A. M.; Galloway, T. S. A small-scale, portable method for extracting microplastics from marine sediments. *Environ. Pollut.* **2017**, *230* (C), 829–837.
- (140) Oßmann, B.; Sarau, G.; Schmitt, S.; Holtmannspötter, H.; Christiansen, S.; Dicke, W. Development of an optimal filter substrate for the identification of small microplastic particles in food by micro-Raman spectroscopy. *Anal. Bioanal. Chem.* **2017**, *409* (16), 4099–4109.
- (141) Tagg, A. S.; Sapp, M.; Harrison, J. P.; Ojeda, J. J. Identification and Quantification of Microplastics in Wastewater Using Focal Plane Array-Based Reflectance Micro-FT-IR Imaging. *Anal. Chem.* **2015**, *87* (12), 6032.
- (142) Bianco, V.; Memmolo, P.; Carcagni, P.; Merola, F.; Paturzo, M.; Distante, C.; Ferraro, P., Microplastic Identification via Holographic Imaging and Machine Learning. *Adv. Intell. Syst* **2020**, 21900153
- (143) Schwaferts, C.; Niessner, R.; Elsner, M.; Ivleva, N. P. Methods for the analysis of submicrometer- and nanoplastic particles in the environment. *TrAC, Trends Anal. Chem.* **2019**, *112*, 52–65.
- (144) Helm, P. A. Improving microplastics source apportionment: a role for microplastic morphology and taxonomy? *Anal. Methods* **2017**, *9* (9), 1328–1331.
- (145) Murphy, M. *Margaret Microplastics Expert Workshop Report*; United States Environmental Protection Agency, 2017.