REVIEW ARTICLE

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A review of methods for measuring microplastics in aquatic environments

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Abstract

An increasing number of reports have been published concerning microplastic (MP) pollution in aquatic environments. Methods used in these studies continue to be updated and lack standardization, so that an up-to-date review pertaining methods for MP research is needed. This critical review examines the analytical methods, including sampling, identification, and quantitation, for MP research. Samples are generally collected from water, sediment, and biota gastrointestinal tract. Manta nets or trawls are prevalently used in surface water sampling, while direct shoveling or box-corer grab are commonly applied in sediment sampling. Microplastics in biota are generally obtained by dissecting organisms and separating livers, gills, and guts. Density separation is frequently chosen to separate MPs from sample matrices. Chemical digestion can dissolve other organic materials and isolate MPs for further identification. Visual sorting should be combined with chemical composition analysis to better identify the polymer type. Pyrolysis or thermal decomposition gas chromatography coupled with mass spectrometry, Fourier transform infrared spectroscopy, and Raman spectroscopy are currently the main technologies for MP identification. Units prevalently used to express MP abundance in water, sediment, and biota are "particles per m^3," "particles per m^2," and "particles per individual," respectively. As MP abundances often varied with the methods used, we recommend that analytical protocols of MPs should better be standardized and optimized. Despite the important progress in analysis of MPs, detection technologies for identifying nano-sized plastic particles are still lacking, and therefore should be developed swiftly.

Introduction

Ever since plastic particles in the sea were first reported in 1972 (Carpenter and Smith 1972), plastic pollution in aquatic environments has drawn increasing worldwide concern (Thompson et al. 2004). In particular, plastic particles,

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designated as "microplastics" in 2004, have been the focus of recent research interests (Thompson et al. 2004). Although there is no agreeable standard, microplastics (MPs) are commonly defined as plastic particles with sizes less than 5 mm (Betts 2009; Cole et al. 2011). Browne et al. (2007) defined MPs as plastic particles with particle size ≤ 1 mm, while those smaller than 100 nm have been dubbed "nanoplastics" (Andrady 2011; Koelmans et al. 2015). Potential sources (primary and secondary sources) of MPs are well documented (Andrady 2011; Cole et al. 2011; Duis and Coors 2016). Primary MPs are created as particulates in commercial products in which they are found, typically facial cleansers (Fendall and Sewell 2009). Other manufactured microscopically sized particles such as in drilling fluids and industrial abrasives are also considered primary MPs (Cole et al. 2011). Fragmentation of large plastic debris through physical, chemical, or biological processes can create smaller plastic particles, which are defined as secondary MPs (Andrady 2011; Cole et al. 2011; Duis and Coors 2016). It has remained a challenge to track the transformation of MPs and accurately

evaluate MP concentrations in environmental matrices. Analytical methods continue to evolve and need to be standardized eventually.

As an emerging environmental contaminant, MPs pose possible risks to the aquatic ecosystem due to their widespread occurrence and potential biological effects (Barnes 2002). The accumulation of MPs in the marine environment typically starts through estuaries, which receive large amounts of plastics from anthropogenic activities (Browne et al. 2010, 2011). Because most MPs are durable and buoyant, they can transport and persist in the environment for long duration and therefore float to remote open waters, where their presence in marine systems is a global concern (Ballent et al. 2012; Cole et al. 2011). Within the marine environment, MPs occurred in marine organisms (Frias et al. 2014; Watts et al. 2014), beach and benthic sediments (Claessens et al. 2011; Stolte et al. 2015; van Cauwenberghe et al. 2013), and water column (Desforges et al. 2014). Ingestion of plastic particles by fish was first reported decades ago (Carpenter et al. 1972). Attention has been paid to the effects of plastic pellets on aquatic organisms, since plastics were reported to be a vector for toxic chemical transportation in marine environment (Browne et al. 2013; Cole and Galloway 2015; Mato et al. 2001). Physical damage (e.g., esophageal blockage) and chemical hazards (e.g., immobilization of *Daphnia magna*) of MPs have been widely investigated (Rehse et al. 2016; Rochman et al. 2013b; Tourinho et al. 2010; Wright et al. 2013b). Sorption of persistent organic pollutants (POPs) (Rios et al. 2007; Van et al. 2012) and metals (Rochman et al. 2014; Turner and Holmes 2015) to MPs may aggravate the toxicology of plastic particles, once ingested (Besseling et al. 2013).

To understand better the distribution and effects of MPs, identification and quantification are the main concerns of most studies to date. Visual sorting and instrumental analysis can yield considerably different results. For example, only 20–70% of MPs identified by visual sorting as plastics are typically confirmed as such by instrumental identification (Lenz et al. 2015). Therefore, inaccurate monitoring methods may greatly under/overestimate MP pollution (Eriksen et al. 2013; Hidalgo-Ruz et al. 2012). A lack of standard protocols for MP sampling and analysis leads to unreliable or incomparable data on MP concentrations and polymer compositions among studies (Hidalgo-Ruz et al. 2012). Therefore, an up-to-date and comprehensive evaluation for analytical methods of MPs in the aquatic environment is necessary, which may be useful in support of establishing criteria for effective monitoring and comparison.

Herein we present a critical review of the most commonly used methodologies for sampling, identifying, and quantifying MPs from various environmental media. Based on an integrated assessment of the utility and limitation of these methodologies, areas for improvements are suggested. Small-sized MPs are the most important for future investigations; hence, MPs larger than 5 mm are not discussed further in this review.

Literature assimilation

An extensive literature review was conducted upon database of Web of Science for publications up to 2017 via searching subjects of "microplastic" OR "microplastics" AND "environment." These literatures were further classified into several topics, such as toxicology, analytical methods for identification and quantification, and environmental matrices (e.g., water, sediment, and biota). The number of publications pertaining MP studies increased rapidly over the last 5 years, especially for analytical methods (Fig. 1).

Field sampling

As the first step of field research, appropriate sampling methods are needed to provide comprehensive and representative MP samples at the locale of interest. Additionally, aerial fallout may be a source of MPs in the environment (Dris et al. 2016); thus, it is suggested that background contamination should be always taken into consideration during the whole study process. Hidalgo-Ruz et al. (2012) summarized three generally recognized sampling approaches: selective, bulk, and volume-reduced sampling methods for sea surface, water column, and sediment. In the present review, updated sampling methods of MPs in environmental matrices (including water, sediment, and biota) were included and the efficiencies of different sampling methods were compared.
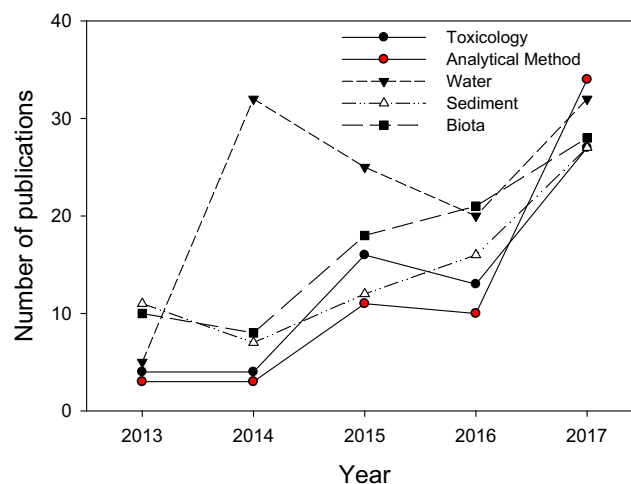

Fig. 1 Literature search results from Web of Science displayed as the number of publications on microplastics from 2013 to 2017, via searching subjects of "microplastic" OR "microplastics" AND "environment." The published papers were further classified into toxicology, analytical methods for identification and quantification and environmental matrices (e.g., water, sediment, and biota)

Water samples

Microplastics are generally buoyant and durable, which allow them to float and persist in water. Surface water sampling is prevalently used by researchers to investigate the occurrence of MPs (Eriksen et al. 2013; Yu et al. 2016; Zhao et al. 2014). There is no standardized or specified depth for water sampling, so various sampling depths are used by researchers (Table S1). For instance, using a rotate drum sampler, Ng and Obbard (2006) detected a large number of MP aggregates in the surface microlayer water of Singapore's marine environment. Song et al. (2014) applied a metal sieve by taping the neuston layers at depth of 150–400 μm, which are typical depths of the microlayer (Cunliffe et al. 2013), to take advantage of surface tension to sample the microlayer water. Water trapped in the mesh of the metal sieve was collected and stored in bottles for further analysis. Using this method, Song et al. (2015b) investigated the distribution of MPs in the seawater microlayer in South Korea, and the MP concentration they detected was much higher than that using other sampling methods (such as trawling and pumping). However, this is a time-consuming method and is more suitable for small-scale surface water sampling rather than for open sea operation.

Given the advantages of large surface areas of open seas and lakes as well as the small volume of final samples, neustonic/manta trawls or nets are prevalently employed for surface water sampling at the depths of 0–0.5 m (Table S1), depending on the submerged height of the trawls used (Anderson et al. 2017; Eriksen et al. 2013; Ivar Do Sul et al. 2014). Almost all surface water samplings with trawls are conducted during research cruises in the sea (Eriksen et al. 2013; Zhang et al. 2017b). The mesh size of trawls ranges from 100 to 500 μm. An obvious technical challenge for trawling is that MPs smaller than the net aperture size may pass through trawls, resulting in underestimation of MP abundance. Because nets with smaller mesh sizes may get clogged with other particulates (e.g., plankton), the most appropriate and widely accepted trawl mesh size for MP sampling is around 300 μm. Particles smaller than 300 μm can be obtained through bulk water sampling using pumps or other techniques (Enders et al. 2015).

Setälä et al. (2016) compared two water sampling methods: trawling (333 μm) and pumping (300 or 100 μm). A higher MP concentration was obtained using a pump with a 100-μm filter than a pump with a 300-μm filter or a trawl. To include plastic samples smaller than a manta trawl mesh size, a submergible pump was used to collect subsurface water at a depth of 4.5 m with a 5-mm aperture size filter (Desforges et al. 2014). They found that size fractions between 100 and 500 μm had the highest MP concentration among all size classes. Similarly, Enders et al. (2015) detected marine MPs down to 10 μm by pumping subsurface water from a depth of 3 m. The greatest number of MPs was observed in the size fraction of 10–20 μm. To prevent clogging, a polytetrafluoroethylene pump was used

by Zhao et al. (2014) to collect bulk water samples at 1 m from the water surface, with further filtration in the laboratory using a 32-μm steel sieve. Pumping water from the subsurface may fail to collect MPs floating on the air-water interface.

Although most MPs are buoyant, high-density plastic particles with additives or other attachments tend to sink into deeper aquatic environments. Hence, MPs in sediment and benthic community are also widely studied (van Cauwenberghe et al. 2013). Taking surface water alone may underestimate the abundance of MPs, although the MP concentration gradient decreases exponentially with increasing water depth (Reisser et al. 2015). Doyle et al. (2011) sampled water at 212 m under the water surface, which required specialized and expensive equipment, but plastic particles (> 0.5 mm) were detected at such depth from only one of the six cruises. To lower the cost for deep-sea MP monitoring, a novel autonomous sensor was developed by Edson and Patterson (2015) for in situ measurement of MPs in the deep water column. This sensor was able to continuously collect seawater from 28 discrete temporal sampling points and measure salinity and temperature at the same time. To improve the reliability of deep-sea sampling, more applicable technologies (e.g., remote sensors) should be developed.

In conclusion, manta trawls or nets are strongly recommended for large-scale surface water sampling in lakes or seas, as they can filter a large quantity of water to collect floating MPs. The aperture size, trawling speed, and sampling time should be standardized, so as to acquire comparable data from different locations. Galgani et al. (2013) recommended a trawl with mesh size of 333 μm and a duration of 30 min for surface seawater sampling. Additionally, a lightweight coarse mesh aluminum cover may strengthen the trawls and prevent them from being broken after a long sampling time. To include MPs sized smaller than the net mesh size, pumping of bulk water is also suggested to complement trawling.

Porous media samples

Porous media can either be sampled for MPs from the surficial layer of coastal/lakeshore beaches or from the seafloor/lake bottom. Because sampling on beaches is easy and convenient, sampling along shorelines has been heavily used worldwide to monitor MP contamination, such as on the coastlines of Singapore (Ng and Obbard 2006), the Tamar Estuary in the UK (Browne et al. 2010), the Belgian coast (Claessens et al. 2011), the German North Sea (Dekiff et al. 2014), Hong Kong (Fok and Cheung 2015), the Bohai Sea (Yu et al. 2016), and even a remote lake in China (Zhang et al. 2016). However, there is no standardized procedure for beach sampling, for which criteria such as tidelines, sampling depth, and sampling volume/area varied (Table 1). Some researchers randomly selected sampling sites on the beach (Nuelle et al. 2014), while

Table 1 Various beach sampling criteria

Parameter	Selection	Reference
Sampling site	High tidal line	de Carvalho and Baptista Neto 2016
	Low tidal line	Harrison et al. 2014
	Whole tidal range	Claessens et al. 2011
Depth	Top 1 cm	Stolte et al. 2015
	Top 2 cm	Martins and Sobral 2011
	Top 3 cm	Browne et al. 2010
	Top 4 cm	Fok and Cheung 2015
	Top 5 cm	Nel and Froneman 2015
	Top 15 cm	Carson et al. 2011
	Top 20 cm	Yu et al. 2016
Area	5×5 cm^2 square	Song et al. 2015a
	15×15 cm^2 square	Mathalon and Hill 2014
	20×20 cm^2 square	Zhang et al. 2016
	25 cm diameter ring	Yu et al. 2016
	1×1 m^2 square	de Carvalho and Baptista Neto 2016

others collected samples by tide (Mathalon and Hill 2014). For the latter, the high strandline or tideline was the most favored beach sampling site. Claessens et al. (2011) demonstrated that the higher water marks might accumulate more MPs than the lower ones.

A stainless steel tool (e.g., shovel and spoon), coupled with latex gloves and cotton clothes, is often used to minimize procedural contamination in beach sampling. The sampling depth varied among studies, with the first 5 cm being the most common (Hidalgo-Ruz et al. 2012). A quadrat (sampling area) of typically but not always 25 cm^2 (Table 1) is frequently used. Either many such squares are sampled, evenly dispersed along the coastline (Van et al. 2012), or a few representative quadrats are selected (Liebezeit and Dubaish 2012). While bulk sampling of beaches may better reflect the occurrence of MPs than representative squares, the workload to characterize bulk samples can be enormous (Nel and Froneman 2015).

With accumulation of biofilms, floating low-density MPs will start to sink, and may end up at the seafloor/lake bottom sediment along with high-density MPs. Woodall et al. (2014) reported that the deep sea might be a major sink for plastics. For sediment sampling, the most frequently used tool is box-corer grab (Vianello et al. 2013), and petite Ponar grabs also have been used in rivers (Castañeda et al. 2014). Superficial (0–10 cm) sediments are generally collected in freshwater, and the top sediment layer (~ 10 cm) is collected from the continental shelf, at a depth of 3500 m (Woodall et al. 2014). A sediment core with a surface area of 25 cm^2 was collected and the top 1 cm was separated for further analysis of MPs (van Cauwenberghe et al. 2013). Turner and Holmes (2015) sampled the top 10 cm of a river sediment core representing sediment accumulation of approximately the most recent 10 years,

as radiometric chronology indicated sediment accumulation rates of 0.8–1.6 cm year^{-1}.

With constant deposition of particulate matter and fluctuation of near-bottom and pore water, MPs can be accumulated in sediment with depth variability. Thus, stratified sediment samples collected with cores may be used to reconstruct the depositional history of MPs (Hidalgo-Ruz et al. 2012). Carson et al. (2011) reported that the top 15 cm accounted for 95% of the total detected MPs, with more than half in the top 5 cm. The lack of a standardized protocol for sediment sampling would result in difficulties in comparing studies using different sampling methods and parameters. In addition, the geochronology of sediment should be taken into account when considering the sediment sampling depth, because MPs in sediment can be transported by bioturbation (e.g., activity of crabs, worms, and other benthic organisms). Currently, surface sediment sampling can only obtain data on recent MP contamination, but no information about accumulation patterns and/or potential sources. Future studies on MP abundance in sediment should also provide geochronology from radionuclides, contaminant profiles, or other means, which can indicate the sediment accumulation rate. Besides, to ensure the representability, at least five sample replicates (5 m between replicates) should be collected from the target strandline (Galgani et al. 2013).

Biota samples

As MPs can be ingested by organisms, biota samples can be used to monitor MP contamination in aquatic environments. The most frequently monitored biota are fish (Peters and Bratton 2016), sea turtles (Wedemeyer-Strombel et al. 2015),

seabirds (e.g., fulmars) (van Franeker et al. 2011), bivalves (Li et al. 2015), marine worms (Wright et al. 2013a), and plankton (Setälä et al. 2014). Some biota are sampled from the wild environment, while others are from farmed specimens. In an attempt to determine the effect of the inhabited environment, Davidson and Dudas (2016) found no significant difference between ingested MP abundances in wild versus farmed clams. Generally, biota is dissected to collect MPs from various tissues and organs. Depending on organism size, the digestive tract can be separated and stored for large individuals, while small individuals can be analyzed entirely (Galgani et al. 2013). Unsurprisingly given their recalcitrance, MPs are often found in digestive systems, which are separated for further treatment and extraction.

In addition, it remains a challenge to obtain a large number of biota samples, especially for top predators, which may hinder accurate measurements of MP contamination in organisms. For example, Wedemeyer-Strombel et al. (2015) found no anthropogenic debris in two leatherback sea turtles. Given the small sample number, it is not at all certain that leatherback sea turtles are free of MP contamination. Considering this scenario, foods ingested by organisms that cannot be sampled in large quantity can be used to indicate any potential MP contamination.

Separation and purification

Flotation

Density separation is widely used to isolate low-density particles from higher-density sand, mud, sediment, and other sample matrices (Dillon 1964). Many MPs, such as polypropylene (PP) and polyethylene (PE), have lower densities than sea water (~ 1.10 g cm^{-3}). High-density plastics, e.g., polyvinyl chloride (PVC), have densities up to 1.40 g cm^{-3} or greater depending on additives and attached biofilms. Various high-density solutions have been employed to isolate MPs from environmental matrices (Table 2). The most favored solution

is saturated NaCl, which is cheap and nonhazardous and has a density of approximately 1.20 g cm^{-3} (Fries et al. 2013). The drawback of NaCl solution is that some plastic particles (e.g., PVC) with density higher than 1.20 g cm^{-3} may not be completely extracted, even though Browne et al. (2010) extracted high-density microplastics using saturated NaCl solution. To overcome this deficit, Nuelle et al. (2014) developed a two-step method, using the air-induced overflow with NaCl solution for pre-extraction and NaI solution (1.80 g cm^{-3}) for further flotation. Recovery rates ranged from 67% (expended polystyrene, EPS) to 99% (PE) for different plastic classifications (Nuelle et al. 2014). Another widely used flotation solution is ZnCl$_2$ (1.50–1.70 g cm^{-3}), which can extract almost all MPs (Liebezeit and Dubaish 2012) but is relatively toxic. Although a range of solutions are used, general procedures for density separation are similar. Briefly, the sample matrix is mixed with a flotation solution, typically by shaking to homogenize the slurry. This slurry is then allowed to sit for several hours so that denser materials (e.g., sand) are settled. Supernatant factions are normally collected for further analysis. To improve extraction efficiency, the solution above the sediment or all solution is subsequently separated by filtration (Stolte et al. 2015; Zobkov and Esiukova 2017a). Sequential extraction was also suggested to improve efficiency (Hidalgo-Ruz et al. 2012). Besides these density solutions, oil can also be used to extract MPs from environmental samples, and an average recovery of 96.1% (± 1.4%) was reported by Crichton et al. (2017).

Technical instruments have been designed for extraction by density separation. Recovery rates for MPs were up to 98 and 100% using an elutriation column and a Munich Plastic Sediment Separator, respectively (Claessens et al. 2013; Imhof et al. 2012). Following the research by Imhof et al. (2012), Zobkov and Esiukova (2017b) evaluated the efficiency of Munich Plastic Sediment Separator for extracting MPs from bottom sediment in the Baltic Sea, and high efficiencies (97.1 ± 2.6%) were achieved. Additionally, Fuller and Gautam (2016) successfully applied pressurized fluid extraction to extract MPs from other matrices, such as glass beads, soil, and

Table 2 Flotation methods/solutions used in previous studies and recovery rates

Method or solution	Density (g cm^{-3})	Recovery rate	Reference
CaCl$_2$ solution	1.30–1.35	> 50% (sizes 100 μm to 1 mm)	Stolte et al. 2015
Potassium formate solution	1.50	Not specified	Zhang et al. 2016
Sodium polytungstate	1.40	Not specified	Corcoran et al. 2009
ZnCl$_2$ solution	1.60–1.70	Not specified	Liebezeit and Dubaish 2012
NaCl solution	1.20	80–100%	Fries et al. 2013
NaCl and NaI solution	1.20 (NaCl), 1.60 (NaI)	94–98% (spheres), 0–98% (fibers)	Claessens et al. 2013
Air-induced overflow	1.20 (NaCl), 1.80 (NaI)	91–99%	Nuelle et al. 2014
ZnCl$_2$ solution	1.60–1.70 (ZnCl$_2$)	96–100%	Imhof et al. 2012
Pressurized fluid extraction	Solvents	101–111%	Fuller and Gautam 2016

municipal waste. However, the recovery decreased with decreasing plastic particle size, which was 97.9% for 30 μm PS and 93.6% for 10 μm PS particles (Claessens et al. 2013). More efficient instruments or solutions should be developed to achieve better recovery with low cost and environmentally friendly consequences. Because most MPs are hydrophobic, surfactants can be used to separate MPs from water (Shen et al. 2002). A combination of flotation and other assistant extraction methods is suggested for future research.

Sieving and filtration

Generally, stainless steel sieves or glass fiber filters instead of plastic tools are used to minimize procedural contamination, and rinsing is always required after each sieving or filtration. A wet sieving process was suggested by Masura et al. (2015), who poured samples through a 5.6-mm stainless steel mesh sieve and then a 0.3-mm sieve. With this step, target MPs were separated for further sorting and identification. To minimize clogging of sieve apertures, a medium coarse mesh sieve of 1 mm is suggested and typically used (Nuelle et al. 2014, van Cauwenberghe et al. 2013). A 500-μm sieve is often used to obtain size fractions of > 500 and < 500 μm. Such fractionation is reasonable, because MPs in the > 500 μm fraction can be identified visually (Hidalgo-Ruz et al. 2012). Other studies employed fine aperture sieves or filters (e.g., 38, 65, and 63 μm) directly to retain target MPs (Claessens et al. 2011; Nel and Froneman 2015; Vianello et al. 2013). Disadvantages of this sieving method include, but are not limited to, easy blocking of sieve holes, difficulty in obtaining wide-range size fractions, and lengthy sample processing time. To save filtration time, stacks of sieves or filters with different aperture sizes may be used to fractionate MPs. Wu et al. (2016) fractionated paint flakes in sediment into four sizes, i.e., < 30, 30–63, 63–200, and 200–2000 μm. This size fraction method is also suitable for MPs.

Size fractionation has varied in different studies. Lambert and Wagner (2016) divided MP samples into three size fractions with a 1-mm sieve and a filter paper of 10-μm pore size. Mathalon and Hill (2014) sieved MPs smaller than 500 μm into several fractions using a series of sieves (500, 355, 250, 150, 106, and 63 μm), yielding detailed size dependency of MPs. However, more fractions may mean less abundant MPs in each fraction. A standardized series of reasonable size fractions is urgently needed to better compare the occurrence of MPs from the same size fraction. Browne et al. (2010) reported that MPs smaller than 1 mm accounted for 65% of the total detected plastic items in sediment along the estuarine shorelines in the UK. A study on the ingestion of MPs by biota found that MPs with sizes smaller than 250 μm amounted to 17−79% of the total (Li et al. 2016). However, accurately identifying and quantifying MPs remaining on the filter paper remains a challenge.

Purification

Organic matter attached to the surface of MPs in environmental samples should be removed to allow clear identification of the type of plastics. A solution of 30% H_2O_2 has been frequently used for that purpose (Liebezeit and Dubaish 2012; Mathalon and Hill 2014; Stolte et al. 2015; Zhao et al. 2014). Nuelle et al. (2014) demonstrated that 35% H_2O_2 performed better than other solutions (e.g., 30% H_2O_2, 20% HCl, and 20–50% NaOH) in dissolving organic matter, although it could cause color fading on MPs. Enzymatic digestion has also been used to dissolve biogenic matter from environmental MPs (Cole et al. 2014). For biota samples, 10% KOH solution has been applied to isolate MPs from the contents of digestive tracts (Foekema et al. 2013; Zhang et al. 2017a). Collard et al. (2015) also successfully extracted MPs by immersing fish stomach extracts in 9% NaClO and then further treating the filtrate with a mixture of NaClO and 65% HNO_3. To accelerate the digestion process, Roch and Brinker (2017) developed a novel procedure combining NaOH and HNO_3 to yield recovery rates higher than 95% with few changes to the characteristics of MPs isolated. High recovery rates (93 ± 10%) of MPs in the soft tissues of mussels were achieved with an enzyme digestion method (Catarino et al. 2017).

Purification is the key to accurate identification of MPs. Although the effects of these solutions on MP characteristics have been demonstrated, potential influences on organic chemicals affiliated to MPs remain unknown. Future efforts should be directed toward the effects of pretreatment on environmental MPs. Without successful pretreatment, the recovery rate of MPs may be lower than predicted. Multi-step pretreatments are suggested to ensure complete removal of biofilms and other organic materials attached on the surfaces of MPs. For example, proteinase K and H_2O_2 are capable of dissolving biofilms and organic materials, while a mixture of H_2O_2 and $FeSO_4$ in Fenton is often used to oxidize organic compounds (Anderson et al. 2017; Masura et al. 2015). Because MPs are a good sorbent for POPs, Fenton-based treatment would destroy POPs in MPs, undermining any attempt to measure POPs affiliated with MPs. This can be circumvented by cleaning of MPs with distilled water when POPs associated with MPs are measured (Van et al. 2012).

Identification

Visual sorting

After separation and purification, target MPs need to be sorted from the remaining matrix. Large plastics can be sorted out directly, while smaller-sized ones need further observation under a dissection microscope (typically a stereomicroscope) after the samples are dried under cover in an oven, generally at

60 °C (de Carvalho and Baptista Neto 2016; Fok and Cheung 2015; Mathalon and Hill 2014). Potential target MPs are thus magnified, sorted, and counted. Norén (2007) suggested criteria for visual sorting of MPs, while this method is suitable for the sorting of large MPs; thus, it is not always applicable and is necessary to use other instrumental techniques (such as FT-IR and Raman spectroscopy) for further identification (Cheung et al. 2016). Lenz et al. (2015) reported that only 68% of visually sorted particles were MPs after identification by Raman spectroscopy, and the success rates of different particles or fibers were dependent on the particle color (Fig. 2). Fibers are easier to be identified than particles, with higher success rates were obtained for fibers (Lenz et al. 2015).

Visual identification may not be accurate enough in this case and may under- or overestimate the abundance of MPs. Although visual sorting is a time-saving method for enumeration of MPs, more reliable technologies are urgently desirable for evaluating the abundance and chemical composition of MPs (Table S2). Song et al. (2015b) found that more MP particles were observed under FT-IR than using an optical microscope. Thus, the number of plastic particles may vary with different methods, e.g., Pyr-GC/MS, TDS-GC/MS, FT-IR, and Raman spectroscopy.

Pyrolysis-GC/MS and TDS-GC/MS

A key benefit of Pyr-GC/MS is that it can simultaneously analyze both the polymer type and organic additives of MPs. Upon being extracted from environmental matrices, plastic particles are thermally deconstructed before the polymer composition of each particle is determined with GC/MS (Fries et al. 2013). This technique allows only one particle at a time to go through the pyrolysis tube, which is both time-consuming and limited by the aperture size of the tube (< 1 mm). The TDS-GC/MS method has been used in analysis of MP composition in environmental samples (Dümichen et al. 2015; Dekiff et al. 2014). Compared with Pyr-GC/MS, TDS-GC/MS can process larger sample mass and measure more complex matrices (Dümichen et al. 2015).

To identify and quantify trace amounts of MPs in environmental samples with Pyr-GC/MS, Fischer and Scholz-Böttcher (2017) employed characteristic fragment ions from each type of plastic as markers. No pre-sorting was required, and the data were robust as r^2 was 0.86–0.99 for the calibration curve (Fischer and Scholz-Böttcher 2017). However, this approach could yield only the mass of each type of MPs but not the particle number count. FT-IR or Raman spectroscopy may be an alternative for MP determination without destroying target particles. Pyr-GC/MS or TDS-GC/MS may be complementary with FT-IR or Raman spectroscopy for comprehensive analysis of MPs (Fischer and Scholz-Böttcher 2017).

FT-IR spectroscopy

FT-IR and its optimized technologies, such as micro FT-IR (Vianello et al. 2013), attenuated total reflectance (ATR) FT-IR (Cheung et al. 2016), and focal plane array detector-based micro FT-IR imaging (Löder et al. 2015), are also used in MP studies. Harrison et al. (2012) compared reflectance micro FT-IR and ATR FT-IR in analysis of polyethylene MPs. Both showed satisfactory performance in identifying polymer compositions (Harrison et al. 2012). ATR FT-IR was better in obtaining spectra of MPs with irregular shapes than micro FT-IR, but was only suitable for analyzing particles larger than 500 μm (Löder and Gerdts 2015). To deal with this issue, Löder et al. (2015) applied focal plane array-based micro FT-IR imaging to determine MPs in environmental samples. Compared to traditional FT-IR, this technology can detect plastic particles down to 20 μm in size and cover a large filter surface area (> 10 mm diameter) (Löder et al. 2015).

Fig. 2 Percentage of plastics with different colors identified by Raman spectroscopy in the total potential plastics after visually sorting. The percentage is shown as success rate. Data were adopted from Lenz et al. (2015)
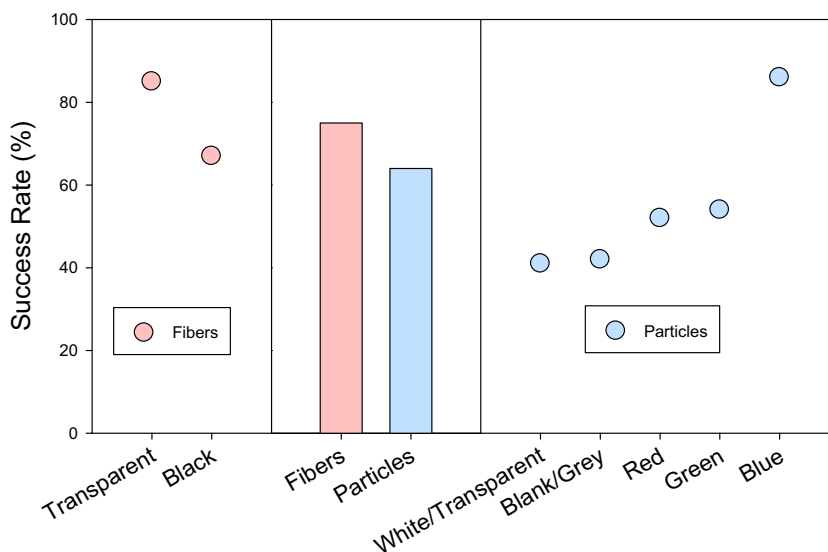
Using FT-IR at a resolution of 8 cm^{-1}, Song et al. (2015a) identified and counted MP particles directly on filter paper after drying at 60 °C. Löder et al. (Löder and Gerdts 2015) suggested that a resolution of 8 cm^{-1} was optimal for micro FT-IR, to produce high-quality data and save measurement time. This resolution was also applied by Vianello et al. (2013) to determine MPs from the bottom sediment of Lagoon of Venice, Italy. Nevertheless, it remains a challenge to apply FT-IR in analyzing ultra-fine plastic particles (e.g., particle sizes < 1 μm) and classifying polymer type from complex environmental samples. Although manual sorting is also required before FT-IR analysis (Claessens et al. 2011), FT-IR is a promising technology but further optimization is required for accurate MP analysis.

Raman spectroscopy

Raman spectroscopy is another promising analytical technique frequently used for MP detection (van Cauwenberghe et al. 2013; Zhang et al. 2016). The main benefits of Raman spectroscopy are that small particles down to 1 μm can be examined and that it has better responses to non-polar plastic functional groups than other analytical methods (Lenz et al. 2015). To minimize false signals with Raman micro-spectrometry, rigorous sample purification is strongly recommended. A suitable wavelength for Raman laser spectroscopy is essential for balancing the enhancement of signal intensity and suppression of sample fluorescence (Löder and Gerdts 2015). In addition, a typical Raman laser spectrometer is much more expensive than a FT-IR, e.g., USD 250,000 versus USD 50,000.

Aside from the three most popular technologies (Pyr-GC/MS or TDS-GC/MS, FT-IR, and Raman spectroscopy), time-of-flight secondary ion mass spectrometry (TOF-SIMS)-based analysis and imaging has also been used for MP detection (Jungnickel et al. 2016). This method can directly detect plastic particles smaller than 10 μm in sea sand with a fragment ion of m/z 113, which showed sufficient power to discriminate polyethylene from environmental matrices (Jungnickel et al. 2016). However, fragment ions from other plastics may overlap with those from environmental matrices, making it difficult to distinguish them. An optimized analytical procedure is still urgently needed to conquer the drawbacks of the methods discussed above, and simultaneously provide comprehensive images of polymer types and numbers in a broad particle size range. Recently, Shim et al. (2016) successfully applied a Nile Red staining method for MP detection in organics-rich samples and achieved a recovery of 98%. This method can also be used for MP quantification following identification (Maes et al. 2017).

Quantification

Quantitative data are needed to illustrate the abundance of MPs in environmental matrices. Upon identification, plastic particles are either counted manually with the assistance of microscope or weighted with a scale. The concentration units of "items/particles per m^2" and "items/particles per m^3" are most commonly used to characterize MPs in surface water sampled by trawling, while MP concentrations in water column or bulk surface water using various aperture size cutoffs are usually quantified as "items/particles per m^3" (Table S2). For sediment samples, sampling quadrate areas are calculated to present MP concentrations as "items/particles per m^2" and "grams/mg per m^2." "items/particles per kilogram dry/wet weight" and "items/particles per m^2 sampling area" are also commonly used for sediment samples. If the shape of sampling area and sampling depth are standardized, sediment MP concentrations may be easily converted from per square meter to per cubic meter. Numbers of plastic particles in sea water (167 ± 138 items per thousand m^3) are typically much smaller than those in nearby estuarine water ($4.14 \pm 2.46 \times 10^3$ items per m^3) (Zhao et al. 2014). As noted previously, an estuarine environment is often impacted by heavy anthropogenic activities from inland (Zhao et al. 2015). To better understand the origins and sources of MPs, more efforts should be directed toward terrestrial environments, as a large portion of land-derived MPs will end up in the oceans. Abundances of MPs ranged from 97 ± 208 items per hundred kg dry sediment in beach sediment of the northern South China Sea (Peng et al. 2017) to 49,600 items per kg dry sediment in the beaches of the East Frisian islands (Liebezeit and Dubaish 2012). When calculated as items per square meter, the number of sediment MP particles ranged from 12 items in beaches of Guanabara Bay of Southeast Brazil (de Carvalho and Baptista Neto 2016) to $13.8 \pm 13.7 \times 10^3$ items in the St. Lawrence River of Canada (Castañeda et al. 2014). These values are much greater than those in surface water, ranging from 5 (0–17) items per km^2 in the northern South China Sea (Zhou et al. 2011) to 0.055–34.2×10^5 items per m^2 of water surface in Xiangxi Bay of the Three Gorges Reservoir in China (Zhang et al. 2017a). Higher MP concentrations in sediment than in water may be resulted from accumulation of MPs in sediment through regular tides. The variability of MP abundances in different regions is derived not only from actual environmental factors (e.g., extent of contamination and population difference) but also from sampling and processing issues such as different filtration pore sizes as previously discussed.

The abundance of MPs may vary with sampling sites, which is not necessarily reflective of the magnitudes of MP pollution, but may also be related to the pore sizes of sampling tools. For instance, sampling in North East Pacific Ocean, Desforges et al. (2014) observed much higher MP abundance using filtration pore size of 0.062 mm than that obtained by Doyle et al. (2011) using a trawl size of 0.5 mm (Table S1). Besides, some studies also collected both coastline beach sediment and adjacent water samples simultaneously. As such, it is not surprising that abundances of MPs were cohesive in

beach and adjacent water samples (Table 3). Discharge and washout from terrestrial environments are most likely the main sources of MPs to the water. Jambeck et al. (2015) estimated that 4.80–12.7 million metric tons of plastic debris entered the oceans from land in 2010. They also predicted that this number might increase tenfold by 2025. It is thus important to monitor the riverine flux of MPs to the oceans, and establishing a uniform and convertible set of metrics to quantify MP abundance in environmental matrices will definitely help.

Microplastics tend to transfer in the aquatic food web (Setälä et al. 2014), as larger biota are shown to accumulate more MPs than smaller ones (Fig. 3). Although they are not in the same food chain, the result can be an indication for bioaccumulation tendency of MPs. The highest average abundance of MPs was found in a whale with 88 items per individual (Lusher et al. 2015), which may be attributed to the large quantity of food in the whale's diet. Approximately 50 fish were found in the digestive tract of this whale, indicating that MPs accumulated in the whale may have partially originated from these fish that may have ingested MPs themselves. Because most plastic wastes are generated inland, discharged to the marine environment, and gradually dispersed to remote oceans, semi-pelagic fish such as the bogue (*Boops boops*) are more closely associated with MPs than pelagic fish (e.g., swordfish), with an average abundance per fish of four items (Nadal et al. 2016) and one item (Romeo et al. 2015), respectively. Because there is no significant correlation between biota weight or size and the number of plastic particles present (Foekema et al. 2013; Tourinho et al. 2010), we suggest the use of items per individual as the unit of MP abundance in biota samples.

Manual counting of MPs is time-consuming, so automatic counting software coupled with identification techniques should be developed for the quantification of MPs. A novel method using Nile Red staining for MP quantification is promoted, which can rapidly detect small MPs (down to 20 μm), and the capability of this method has been demonstrated (Erni-Cassola et al. 2017; Maes et al. 2017). In addition, the units of weight and count are suggested to be standardized or exchangeable for comparison of MP abundances between studies.
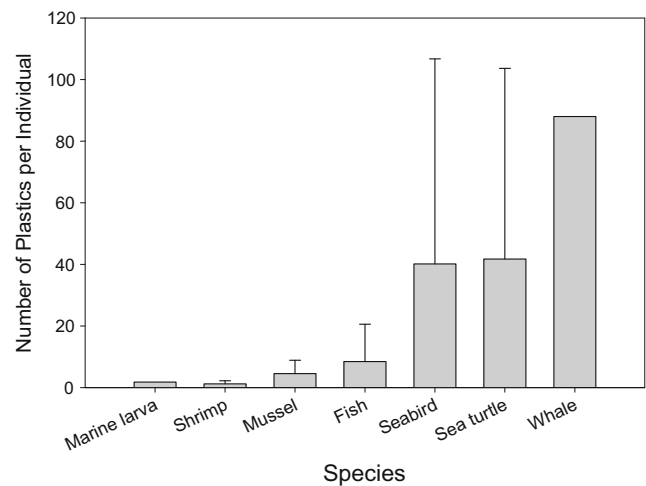

Fig. 3 Summarized numbers (average ± SD) of plastic items per individual organism from literature surveys. Data were adopted from the studies of Kaposi et al. (2014) for marine larvae; Devriese et al. (2015) for shrimp; Li et al. 2015, 2016) for bivalves; Possatto et al. (2011), Romeo et al. (2015), Miranda and de Carvalho-Souza (2016), Nadal et al. (2016), and Peters and Bratton (2016) for fish; Tourinho et al. (2010) and van Franeker et al. (2011) for seabirds; Barnes (2002) and Tourinho et al. (2010) for sea turtles; and Lusher et al. (2015) for a whale

Quality assurance and quantity control

During the whole sample process, quality assurance and quantity control (QA/QC) is essential and of great importance for data accuracy. In field sampling, procedural blanks (containing pure water) and spiked blanks (containing pure water and a certain amount of MPs of known composition and abundance) should be prepared (Galgani et al. 2013). Non-plastic sampling tools, latex gloves, and cotton clothes should be used during sampling. During the extraction process in laboratory, recovery rates of standard MPs (with the similar sizes and classifications of field samples) using the applied extraction method should always be provided (Stolte et al. 2015). Since there are some synthetic fibers in the atmospheric fallout (Dris et al. 2016), blanks and inter-laboratory tests can be performed to minimize the effect of the experimental environment. In addition, Zobkov and Esiukova (2017a) suggested that an internal standard and occasional empty runs can ensure the reliability of extraction efficiency in processing sediment

Table 3 Abundances of microplastics in beach sediment and adjacent water

Location	Microplastics at beach	Microplastics in nearby water	Reference
Brittany, France	1 ± 2 items per kg dry weight	24 ± 35 items per 10^2 m^3	Frère et al. 2017
Three Gorges Reservoir, China	80–864 items per m^2	0.55×10^5–342×10^5 items per m^3	Zhang et al. 2017a
South Africa Coastline	689 ± 348–3308 ± 1449 items per m^2	258 ± 53–1215 ± 277 items per m^3	Nel and Froneman 2015
Northern South China Sea	33 (3–375) items per km^2	5 (0–17) items per km^2	Zhou et al. 2011
Lakes in Italy	112–234 items per kg dry weight	1–5 items per m^3	Fischer et al. 2016

samples. For MP identification, reference data of standard MPs from the instrument data base should be used to compare with field collected MPs (Ng and Obbard 2006). When analyzing MP samples from heavy contaminated sites (more than thousands of particles), it is hard to count all particles one by one. Grouping/pooling is always performed to roughly estimate the total MP concentration, and at least 10% of particles should be instrumentally identified and counted (Mahon et al. 2017). Besides, the percentage of identified MPs in the total suspected particles should also be provided; otherwise, the concentration of MPs would be overestimated.

Recommendations

This review has provided an overview of MP detection in aquatic environments (Fig. 4), and discussed numerous methods used in sampling, extraction, identification, and quantification of MPs. Existing data are often incomparable because of the various approaches used. To better compare MP contamination worldwide, procedures and methodologies should be standardized so that they are practical over the entire analytical process. Amid the limitations of existing methods and technologies that have been discussed in this review, recommendations are given as follows.

a) For sediment sampling, selection of sampling sites is of high importance to provide truly representative MP contamination in the target location. The sampling depth and location (e.g., high, mid, or low tide line) should be specified and standardized for sediment sampling.

b) Manta trawls or nets are strongly recommended for large-scale surface water sampling in lakes or seas, since they can filter a large quantity of water to collect floating MPs during sampling. To include size range of MPs smaller than the net mesh size, pumping of bulk water is also suggested to complement trawling.

c) For biota sampling, a certain food web of samples should be better collected, as done in chemical contaminant studies (Kidd et al. 1995; Xu et al. 2014).

d) Density separation is still strongly recommended to float MPs from sediment samples. Technologies (e.g., separator) based on density separation or assistant reagents (e.g., surfactants) complementary to density separation should be promoted.

e) During the purification stage, 30% H_2O_2 solution combined with $FeSO_4$ (Fenton) is highly recommended to remove organic matter. The effects of purification solutions on other target organic compounds should be fully investigated in future research.

f) It remains a challenge to identify plastic particles with sizes smaller than 1 μm. This is the limit of a visual microscope and is necessary for sorting purposes before instrumental identification. Complementary technologies, such as X-ray diffraction, X-ray photoelectron spectroscopy, and energy disperse spectroscopy, are encouraged to be used for small particle identification.

g) Manual counting is generally used for MP quantification; however, this method is time-consuming. Automated quantification technology (e.g., flow cytometry and imaging technologies coupled with statistical analysis) could be adapted to efficiently obtain the concentrations of MPs in environmental samples. Dynamic light scattering is

Fig. 4 Flow chart of analytical processes for analyzing microplastics in various environmental matrices
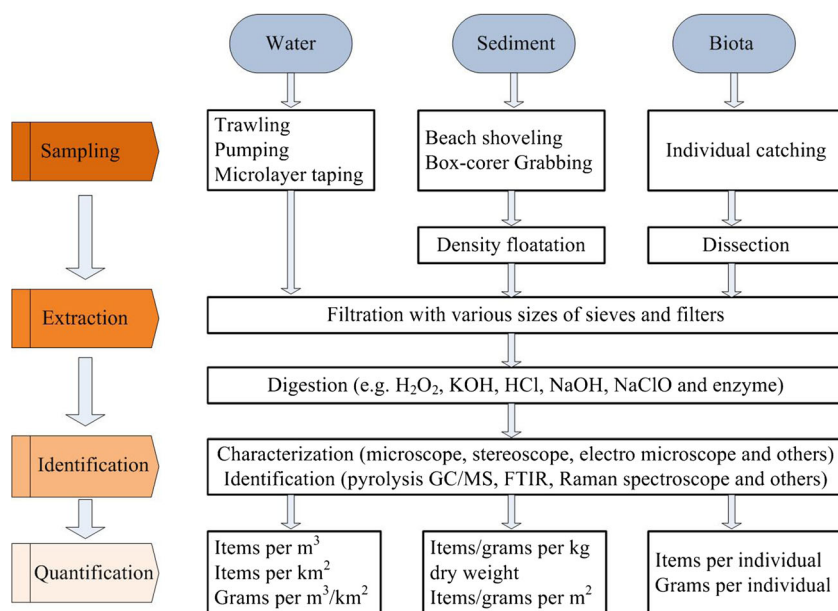
suggested to provide the size distribution of MPs, although nano-sized MPs will be difficult to detect using this technique against the scattering provided by much larger MPs.

Given the current knowledge discussed in this review, efficient detection methods could become a solid foundation for research to understand MP distributions worldwide. Because plastic wastes have been classified as hazardous materials (Rochman et al. 2013a), policies should be built to better manage plastic pollution (Tibbetts 2015). This would rely on the availability of more comparable and robust data on the distribution, fate, transport, and effects of MPs in the marine environment. With such efforts, contamination of MPs may be reduced or eliminated.

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