Meta-analysis of Error Sources in the Determination of Micro- and Nanoplastics

by

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ABSTRACT

The occurrence of micro-and nanoplastic (MNP) debris in the environment is a research area of considerable public health concern. Various combinations of methods for extraction, isolation, and quantification of MNP have been applied but literature studies evaluating the appropriateness and efficacy of these protocols are lacking. A meta-analysis of the literature (n=134; years 2010-2017) was conducted to inventory and assess the appropriateness of methodologies employed. Some 30.6% of studies employed visual identification only, which carried a calculated misidentification error of 25.8-74.2%. An additional 6.7% of studies reported counts for particles smaller than the cutoff value of the selected collection pore size, and 9.7% of studies utilized extraction solution densities which exclude some of the polymers commonly occurring in the environments investigated. A composite value of data vulnerability of 43.3% was determined for the sample, indicating considerable weaknesses in the robustness of information available on MNP occurrence and type. Additionally, the oxidizing solutions documented in the literature frequently were deemed unsuccessful in removing interfering organic matter. Whereas nanoplastics measuring <1 μm in diameter are likely principal drivers of health risk, polymer fragments reported on in the literature are much larger, measuring 10+ μm in diameter due to lack of standardized methods. Thus, current inventories of MNP in the environmental MNP feature data quality concerns that should be addressed moving forward by using more robust and standardized techniques for sampling, processing and polymer identification to improve data quality and avoid the risk of misclassification.

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Introduction

Beginning in 1972, microplastic pollution has been documented in every part of the world from ice cores in the Arctic to polymeric particles in Antarctica (Munari et al., 2017; Obbard et al., 2014; Lusher, Tirelli, O'Conner, & Officer, 2015; Cózar et al., 2017; Bergmann et al., 2017). Despite the pervasive nature of polymeric litter, uniform methods to analyze and quantify these microplastics have yet to be widely adopted. After more than four decades of polymeric litter studies, methods of quantification are finally beginning to approach the smaller size range: nanoplastics (Catarino, Macchia, Sanderson, Thompson & Henry, 2018).

Toxicological studies on the burden of microplastics taken up by biota have shown microplastics <110 µm enter the blood stream and are transported into the lymphatic system, bile, urine, and cerebrospinal fluid. Accumulation occurs in the liver, kidney, and gut at sizes <20 µm of mice, producing neurotoxic responses among others (Deng, Zhang, Lemos, & Ren, 2017). Microfibers have even been discovered at sizes up to 135 µm in the lungs of lung cancer patients with links to carcinogenic effects at sizes >10 µm in length (Pauly et al., 1998; Omenn et al., 1986; Wright & Kelly, 2017). Moreover, 78% of the US EPA's priority pollutants are utilized to either synthesize plastics or have been found adsorbed onto plastic litter (Rochman et al., 2013). Sources of microplastic ingestion include bottled water, sea salt, sugar, beer, honey, shellfish, and crustaceans (Liebezeit & Liebezeit, 2017; Schymanski, Goldbeck, Humpf, & Fürst, 2018; Yang et al., 2015).

One common polymer identification classification approach focuses on the size of plastic debris. The largest plastics occur as macroplastics (>25 mm) and mesoplastics (5-

25 mm). Although microplastics (MP) are sometimes referred to as particles less than 5 mm, academic literature is establishing a lower size limit of 1 µm along the longest axis (Andrady, 2011). Particles measuring <1 µm are classified as nanoplastics (NP) (Mattson, Jocic & Doverbratt, 2018). However, upper and lower size limits are openly debated in literature (Cole, Lindeque, Halsband, & Galloway, 2011).

Another sub-classification scheme distinguishes primary and secondary micro-and nanoplastics (MNPs), dependent upon their occurrence in a manufactured, virgin state or as the progeny of primary plastics, resulting from mechanical, chemical, thermal, and ultraviolet degradation. Thus, both primary and secondary plastics over time do break down along a continuum from macro- to meso- to micro- to nanoplastics.

Due to polymer's ubiquitous applications, MNPs are documented to occur from a variety of merchandise as well as from releases from point and non-point sources. Sources of primary MNPs include but are not limited to facial products, toothpaste, glitter, gel nail polishes, abrasive paints, manufactured pellets, and engineered nanomaterials. Sources of secondary plastics, as previously mentioned, include the plethora of plastic debris in the environment breaking down such as macro-and mesoplastic merchandises. These macro-and mesoplastics include synthetic clothing, food containers, wrappers, utensils, plastic bags, and fishing supplies. Point sources of MNPs into the environment include wastewater treatment plants, city dust, and other highly anthropogenic locations produced from abrasion of macroplastics.

MNP composition differs by environmental sample type and location. A spectrum of combinations of organic polymer additives, inorganic polymer additives, and polymer types are documented. The most common microplastic litter in sediment is as follows:

polyethylene (PE), polypropylene (PP), polystyrene (PS), polyethylene terephthalate (PET), polyvinyl alcohol (PVA), polyamide (PA), polyurethane (PU), and polyvinyl chloride (PVC) (Burns & Boxall, 2018). Standard thermoplastics make up the vast majority of plastics produced and account for approximately 229 million metric tons globally annually (Plastic Europe, 2015). Of the polymers listed, monomer constituents have well established toxicity including PVC and PU causing carcinogenic effects and cellular mutagenicity (Lithner, Larrson, & Dave, 2011).

Organic plastic additives such as diethyl phthalate, diethylhexyl phthalate (DEHP), disobutyl phthalate (DBP), and dimethyl phthalate are recorded in environmental microplastic samples as well (Fries et al., 2013). Inorganic plastic additives found in MPs include aluminum, titanium dioxide, barium, sulphur, oxygen, and zinc (Fries et. al, 2013). Notably, DEHP and DBP have been correlated with serious developmental issues most likely due to endocrine disrupting effects (Heudorf, Bolker, & Jürgen, 2007).

Currently, MP sample processing consists of three phases: collection, extraction, and analysis. The collection phase can be bulk removal in sediment, water, or ice whereas size-specific removal involves various pore sizes of nets, sieves, or filters. Next, extraction involves either density separation (DS), oxidation, DS and oxidation, or no additional processing (NAP). The analysis phase then involves either visual only, material only, visual and material, or visual sample interrogation.

In the absence of a uniform method, this study aims at assessing the certain, or robust data, produced by historical methods. Likewise, addressing the uncertain, or vulnerable data, produced by other methods will provide additional clarity. The objective of this study was to identify vulnerable or flawed microplastic studies in order to prevent

their propagation in future studies. Additionally, recognizing potential sources of error in microplastic analyses will assist in creating methods for environmental nanoplastic analysis and quantification as literature progresses to this size range.

Methods

Literature Search. A comprehensive database of environmental microplastic studies was acquired following PRISMA meta-analyses guidelines (McInnes et al., 2018) of the following databases: Google Scholar, Science Direct, Arizona State University's Library One, and ResearchGate. Advanced search terms included small plastic litter (microplastic, nanoplastic, microfiber, or plastic litter) and matrices of occurrence (water, wastewater, sediment, or soil). Journal articles excluded were as follows (1) a comparison of methods or novel methods, (2) occurrence of MNPs within organisms, (3) unpublished, (4) not accessible within the databases listed above, or (5) not translated into English. Inclusion criteria focused on articles which target the microplastic size range and collect, extract, and analyze these particles within the study. A sample search term of Google Scholar is as follows: 'allintitle: MICROPLASTIC -METHOD -MICROALGAE -FOOD -BIOACCUMULATION -PREDICTION -MODEL -PLANKTON -BIOTA -HERRING -ALGAE -GASTROINTESTINAL -COD -GUT -TROPHIC -MUSSEL -MUSSELS -FISH -CRAB -MODEL -CRUSTACEANS -INGESTION' (year: 2010-2017) (n=288); the '-' term denotes NOT. Studies were screened individually to further identify their suitability. Exclusion criteria included papers exploring novel identification methods and those which enumerate microplastics which occur within biota. Additionally, abstracts were reviewed to further apply

exclusion and inclusion criteria. The literature search considered publications occurring on or before January 2018. The final sample of studies entering the analysis was n=134.

Data Extraction and Meta-Analysis. Literature which met all criteria were then individually inventoried for multiple parameters including: sample environment, minimum collection, filter or sieve processing, and particle size, oxidative solution(s), density solution(s), applied quality control method(s), applied analysis method(s), and spectroscopy library reference for material identification. Oxidative solutions were categorized as none, wet peroxide oxidation (WPO), WPO & other, undefined (ND), and other. Density separation solution(s) were categorized as NaCl, ZnCl, none, ND, and other. Applied quality control methods include the following: avoidance of synthetic instrumentation or dishware, procedural control blanks, extraction control blanks, and nonsynthetic clothing, e.g. lab coat. Applied analysis methods were then categorized into types including: visual only, visual and macro-spectroscopy [Fourier Transform Infrared Spectroscopy (FTIR); Raman], micro-spectroscopy (µ-FTIR; µ-Raman; Scanning Electron Microscope-Energy Dispersive X-ray Spectroscopy (SEM-EDS)], visual and pyrolysis gas-chromatography mass spectrometry (Pyro. GC-MS), and other. For samples which apply spectroscopy, material identification reference are differentiated by manual with reference, manual with no reference, and automatic referencing which calculates a percentage match or best fit to material. Simple statistical analyses were then conducted within Microsoft Excel to innumerate and sort each category. All data included in the meta-analysis are provided in the Supporting Information (SI).

Error analyses were conducted from this literature sample to determine sources and quantify prevalence. Data to determine particles analyzed outside of the sampling size range was extracted from the text of literature as well as graphical images. Sizes were determined from graphical images using microscope scale bars when provided. All meta-analysis literature which contained author's calculated error values for misidentification of organic particles for microplastics were collected alongside total particles analyzed for the cumulative error analysis. Density outliers were isolated through binning data.

Identification Error Analysis. A weighted average of cumulative error was then calculated using Equation 1.

$$Cumulative\ Error = \frac{\sum_{i=1}^{n}(\%\ error\ \times\ n)}{\sum_{i=0}^{n} n} \quad \text{Eq. 1}$$

Where: $\%\ error$ is percent error of the respective study.

n is the number of particles analyzed.

Composite Data Vulnerability Analysis. The number of studies which all error sources occur in literature were combined to determine one composite value. Any repeated error sources were omitted.

$$Data\ Vulnerability = O(M) + O(D) + O(P) - O(R) \quad \text{Eq. 2}$$

Where: $O(M)$ is the occurrence of misidentification.

$O(D)$ is the occurrence of improperly applied density separation.

$O(P)$ is the occurrence of quantifying particle smaller than the minimum pore size.

$O(R)$ is the occurrence of repeated error sources.

Results

Literature Meta-Analysis Results. Upon literature search (n=380) and application of exclusion criteria, a total of 134 studies published between 2010 and 2017 remained for consideration. Among this body of literature, studies enumerate MPs from sediment, saltwater, freshwater, brackish water, wastewater treatment plants (WWTPs), street dust, and sea ice. With respect to sample pre-treatment, a majority of the literature applied no oxidation (66.4%) (n=134*)* or wet peroxide oxidation (WPO) (26%)as well as the following: (other, 3.7%; WPO and other, 2.2%; undefined, 1.5%). Solutions applied for density separation included primarily NaCl (41.8%) followed by additional separation compounds or lack thereof (none, 35.8%; undefined, 9%; other, 8.2%; ZnCl, 5.2%). None dictates the process was never applied. Undefined solvents are essentially studies which state using the process but never identify the solvent. Quality control methods applied include none (40.2%) followed by one (22.4%), two (20.1%), three (14.9%), and four (2.2%). Methods of polymer identification were also analyzed with the majority applying visual and macro-spectroscopy (34.8%), visual only (30.6%), micro-spectroscopy (13.4%), other (9.7%) spectroscopy only (5.2%), and visual and Pyro. GC-MS (2.2%). The "Other" data category in Fig. 1 includes combinations of methods that add a level of robustness to the study design: Pyrolysis Gas Chromatography-Mass Spectrometry (Pyro. GC-MS) and microscopy with Fourier Transform Infrared Spectroscopy (FTIR), 0.7%; microscopy, FTIR, and differential scanning calorimeter (DSC), 0.7%; microscopy and DSC, 0.7%; Pyrolysis Gas Chromatography-Mass Spectrometry (Pyro. GC-MS) and

Scanning Electron Microscopy- Energy Dispersive X-ray Spectroscopy (SEM-EDS), 0.7%; microscopy, FTIR, and GC-MS, 0.7%; microscopy, Raman, and FTIR, 0.7%; microscopy, micro-FTIR, and Attenuated Total Reflection (ATR)-FTIR, 0.7%]. Of studies which included spectroscopy, 26.5% verify polymer identity with no reference library; 39.8% manually verify polymer identity with a reference library; 33.7% employ automated identification libraries that include best fit or percent match.

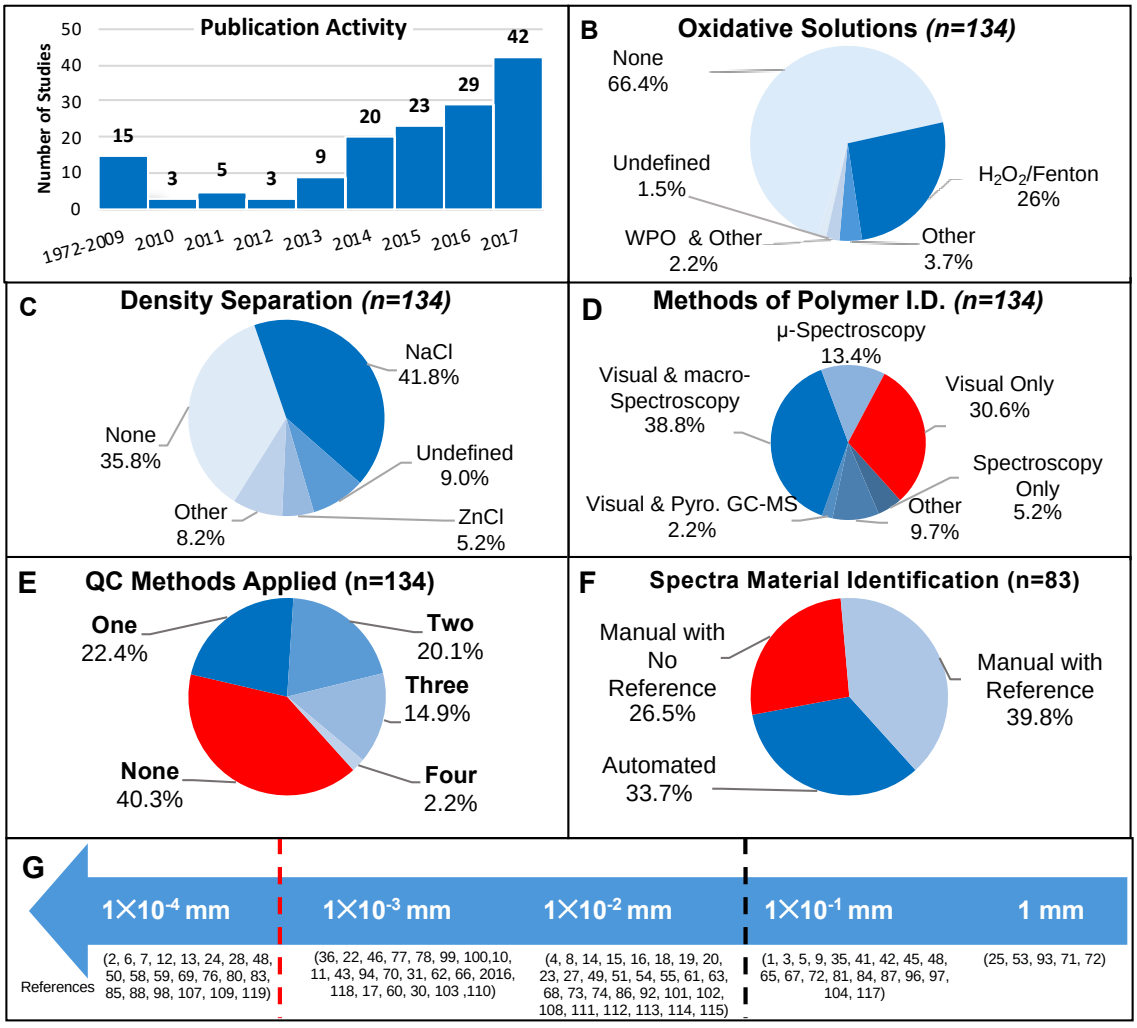

Figure 1. QC=Quality Control. Results from the meta-analysis on microplastics literature (n=134). Number of peer-reviewed publications by year fitting the exclusion criteria of this review (A), pre-processing of environmental samples (B, C), applied characterization or visual methods to sample (D), and smallest sieve, filter, or mesh size applied during processing. The red sector highlights lack of any spectroscopy, spectrometry, or other material verification instrument. Applied quality control methods includes a combination of sample contamination mitigation techniques such as nonsynthetic lab instruments and apparel (E). For studies which utilizes spectroscopy, a

differentiation of manual, manual no reference, and manual with reference material identification libraries are shown (F). The red and black dashed lines show the average smallest extraction pore size of the sample compared the average smallest particle studied (F). 'None' dictates the process was never applied. Studies which utilize the processing method but do not identify the solvent are shown as 'undefined'. The other category includes rarely applied methods.

Vulnerability to Potential Misquantification.

Appropriateness of sample screening procedure. Aqueous and sediment sample extraction typically varies in that aqueous samples focus on the filter cake whereas sediment samples focus on the filtrate. Thus, incidental capture and enumeration of particles in screens, i.e. nets or sieves, has a false positive effect on aqueous samples and false negative effect on sediment samples. Of course, false negatives in this instance would not be recorded; however, false positives are noted via microscope images of samples, description of minimum particle size counted, and other ways. This source of false positive occurs when applying sieve, mesh, or nets to retain particles of analysis and these incidentally captured particles by pore blockage. A small portion (6.7%;n=134*)* of literature analyzed particles which were smaller than the retaining pore size. Mismatches of collection size and pore size are summarized in Table 1. All of these studies occurred in aqueous samples.

Particle Size Targeted in Collection (mm)	Smallest Particle Size Enumerated (mm)
≥ 0.153	0.063
≥ 2	0.05
≥ 0.35	0.3
≥ 0.35	0.3
≥ 0.3	0.1
≥ 0.35	0.3
≥ 0.33	0.32
≥ 0.15	0.1
≥ 0.33	0.24

Table 1. Mismatch of particles analyzed and collection pore size. Each row is from a different study. A total 6.7% of the meta-analysis literature database quantifies these particles which are incidentally captured.

Appropriateness of density separation solution. A range of solution densities are applied for polymeric separation; however, some applied densities do not include all environmental microplastic densities in literature (Figure 2). Binned solute densities were analyzed with respect to previously discovered microplastic polymer densities. The range of density (ρ) least effective at isolating common polymers was found to be in the range $1.3\ g/cm^3 \geq \rho \geq 1.14\ g/cm^3$ (Figure 2). Solutes within this binning range composed 9.7% of the 13 studies. Of all studies which apply density separation, 15.1% excluded some materials (n=86) However, some studies focus upon one layer of the water column, i.e., sea surface microlayer, littoral zone, deep-sea, and subsurface studies, and may assume an exclusion of microplastic densities simply by sample environment, disallowing for capture of transient plastics moving through the respective environment.

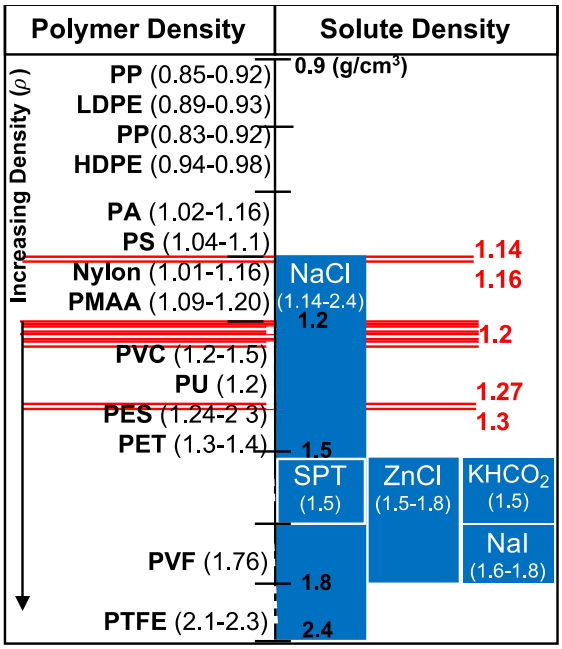

Figure 2. Densities of common marine polymers and densities of applied solutions for separation in 134 studies. Red lines denote the occurrence of densities found in literature that exclude common polymeric materials. The empirical density used is shown to the right of each line. An error occurs when $\rho_{polymer} < \rho_{solute}$.

Misidentification. In terms of microplastic analysis, robust data is preferred to verify polymeric composition. If the material composition is not defined, then the particle may not be polymeric. As described in Figure 1.D, 30.6% of literature categorized particles as microplastics with only microscopy. Without the application of material identification, an identification error of 25.8-74.2% (n=6) was determined utilizing recorded errors of visually identified microplastics. This is also sometimes referred to in literature as observer bias and is one large source of error preventing equal comparison across microplastic literature. Particles commonly mistaken for microplastics or microfibers include fly ash, cotton fibers, and red algal fibers (Dubaish & Leibezeit,

2013) . Additionally, misidentification potential is found in studies categorized as “other” (mechanical testing, 0.7%; “hot needle testing”, 0.7%; strong acids, 0.7%). Mechanical testing is described as physical pressure applied to the material. Hot needle testing applies heat to a potential synthetic material to verify by melting point; strong acid tests also determined if synthetic material melted when exposed.

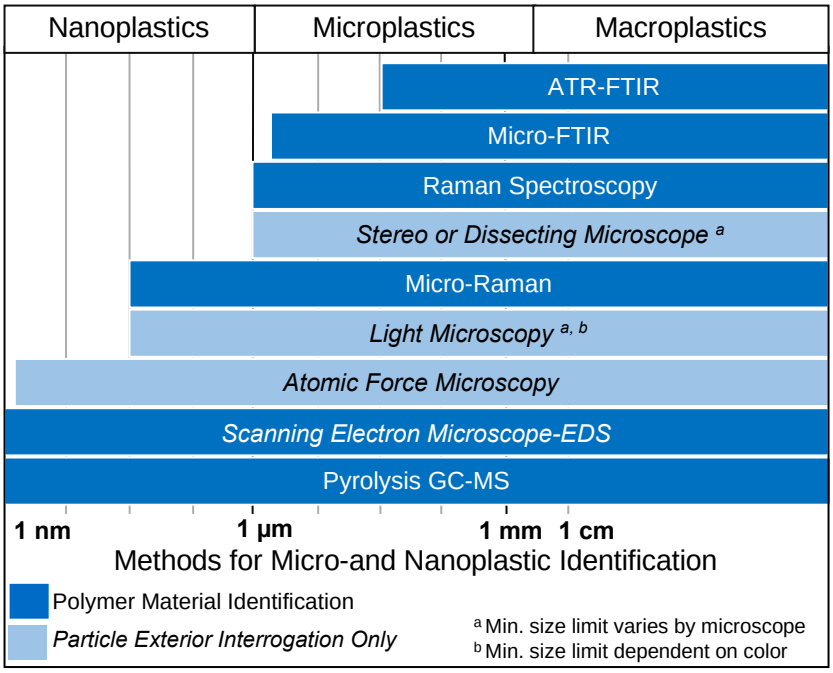

Figure 3. Characterization of micro-and nanoplastics by size (Ref. in Appendix D).

Other Sources of Misquantification.

Contamination of laboratory and sampling environments is a well-documented occurrence with varying effect on quantification (Mai et al., 2017). Contamination mitigation methods include utilizing non-synthetic materials for all processes including lab coat (1), instruments (2), sieves (2), dishware (2), and more. Perhaps the most robust

of all would be the subtraction of both extraction (3) and procedural blanks (4). In the peer reviewed studies analyzed, 40.3% included none of the four contamination mitigation techniques numbered above. Moreover, only 2.2% applied all four mitigation techniques. Naturally, cleansing of instruments, filtering solvents, utilizing a fume hood, and air filtration devices further prevent contamination. Lastly, utilization of controls or processing blanks, sampling blanks, testing bench surfaces, identification of research vessels color and material ID (Bagaev, Khatmullina, & Chubarenko, 2017), and more methods can prevent the contamination of samples.

Importance of Nanoplastic Analysis. The smallest particle size analyzed from an environmental matrix is currently ~10 µm; however, the presence and potential abundance of nanoplastics are widely acknowledged to be produced from degradation as well as occurring in a manufactured state. Although the average minimum pore size of filtration or screening in the meta-analysis is 146.8 µm, data shows that the average particle size included in quantification is 354.8 µm (n=134). Additionally, an increasing presence of decreasing particle size with continued exposure happens over relatively short time periods (Hahladakis et al., 2017).

Moreover, microplastic toxicity is directly related to particle size and mechanism of exposure. Persistent microfibers have been discovered in lung tissue of lung cancer patients at sizes up to 135 µm. Beginning at 130 µm, particles begin to enter the lymphatic system. Studies have also noted the occurrence of microplastics at up to 110 µm in blood, urine, and cerebrospinal fluid of dogs following ingestion. Moreover, deposition in the liver, kidney, and gut increase for particles <20 µm (Deng, Zhang,

Lemos, & Ren, 2017). Overall, particles size influences the transport, residence, toxicity, and other major factors of microplastic toxicity.

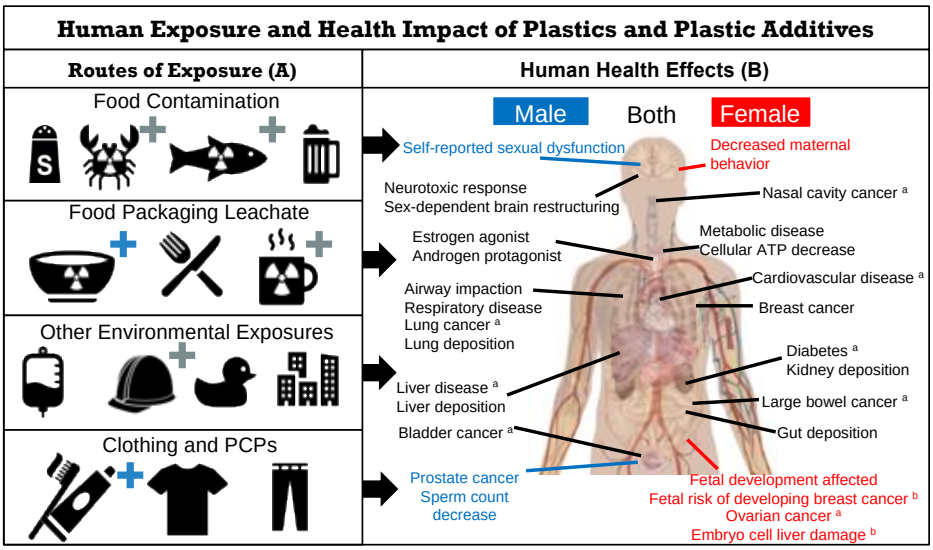

Figure 4. Health impacts of plastics and plastic additives. Gray arrows denote sources of exposure which have not been regulated in the US. Blue arrows denote exposure which have regulations in place (A). Anatomy Graphic: By Mikael Häggström (All used images are in public domain.) [CC0], via Wikimedia Commons (B).

Interestingly, cases which involve joint replacement with polymeric materials also exhibit localized inflammation, necrosis, and redistribution of particles to the lungs. Approximately 14% of human patients with polymeric joints were discovered to have nanoplastics deposited in the liver or spleen. This is consistent with other studies which conclude particles 0.5-50 µm cause a foreign body response (Wright & Kelly, 2017).

Discussion

This literature review identified a number of vulnerabilities in the collection, extraction, and analysis of environmental MNP. All in all, very few microplastic methods of extraction, isolation, and quantification are similar. Error sources begin with sample processing as 6.7% of studies analyzed particles retained in nets, sieves, or filters smaller than the pore size of the extraction instrument. Next, 50.4% of studies used only density separation with 9.7% of all studies densities leaving out portions of the environmental polymer population. Furthermore, oxidation is not always successful for removing organic matter according to literature. No processing, e.g. density separation or oxidation, remains the greatest risk for sample misidentification particularly if applied in conjunction with only visual identification. Most importantly, 30.6% of the sample applied no methods of material identification to verify polymeric composition. Within 30.6% of meta-analysis studies, an average 25.8-74.2% of misidentified microplastics was calculated from published data sources. Some studies binned within the "other" category contain error sources through polymeric analysis involving vulnerable methods such as 'flame tests' and mechanical prodding of objects accounting for 1.4%.

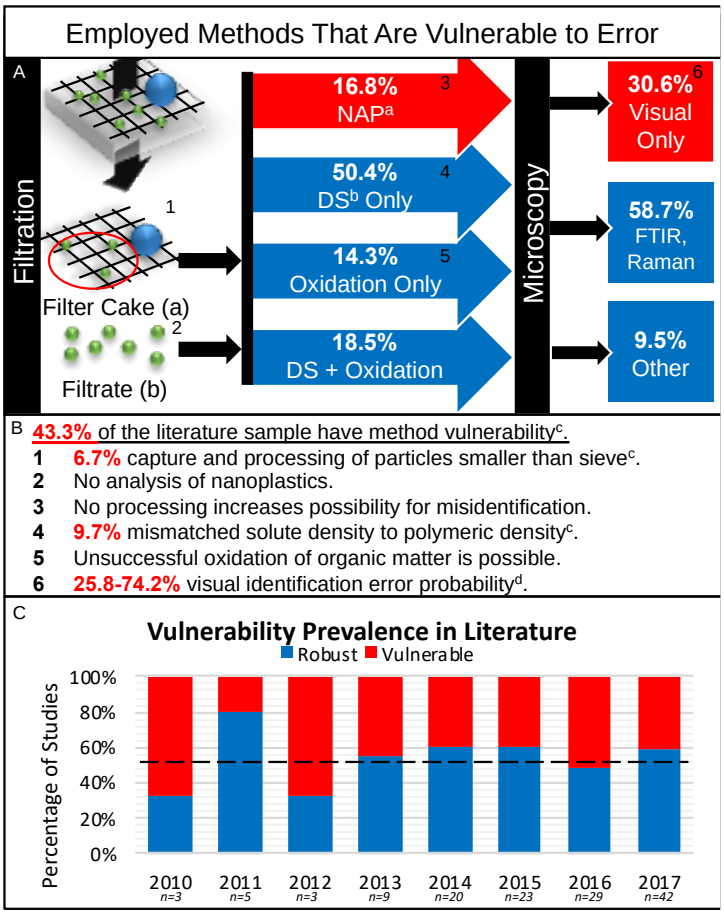

Figure 5. Schematic illustrating potential source of error and recommended method. [a] NAP=No Additional Processing. [b] DS=density separation. [c] *n*=134. [d] *n*=6. Path (a) illustrates sampling practices which focus on the filter cake or filtrate, respectively. The blue boxes denote robust methods for analysis while red boxes denote vulnerable methods (A). A ranking of potential for error source is shown (B). Despite a positive trend in publication activity, no trend can be discerned for method vulnerability. Data vulnerability (43.3%; *n*=6) is shown with the black dashed line (C).

Prospectively, studies can avoid these sources of error through design of methods focusing on the particular sample composition and desired size range. Mismatch of particle size and pore size is easily avoided with extraction techniques which contain a larger range of microplastics such as bulk or core samples. Additionally, binning particles by size is a reliable way to prevent any particles beyond the size range of sampling.

Optimal density separation is applied as a two-stage method of a hyper-dense solution followed by a moderately dense solution ($2.0\ g/cm^3 \leq \rho_1$ then $1.3\ g/cm^3 < \rho_2 < 1.8\ g/cm^3$). Note the solution is soaked for several hours, the supernatant is extracted carefully, and then the remaining solution is sonicated and separated by density two additional times (Mai et al., 2017). Oxidative solutions are optimized as a multi-stage system to target a desired level of organic matter removal. These solutions must be cleansed, sonicated, and filtered between stages as well. Application of density separation is most beneficial in samples consisting of sediment, whereas oxidation techniques would be most beneficial in highly organic sample environments such as saltwater. Both density separation and oxidation would be optimally applied to a highly organic sample such as wastewater.

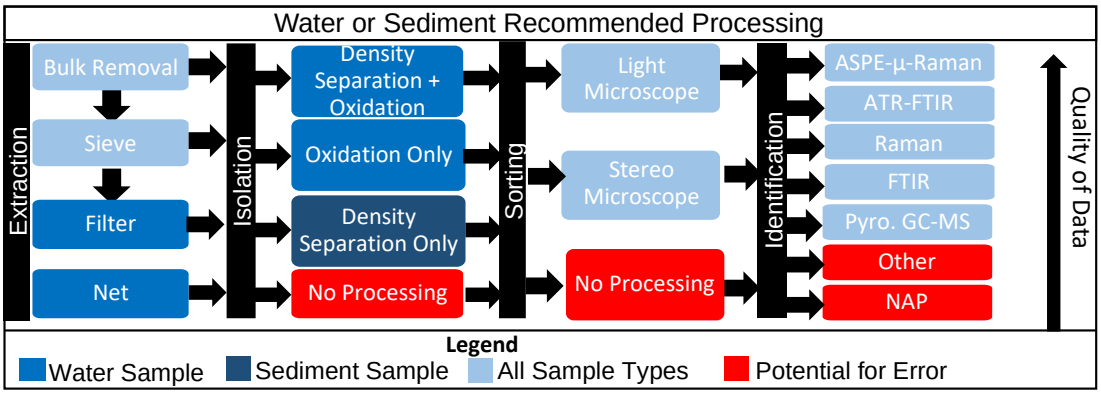

Figure 6. Recommended method diagram with increasing data quality.

Due to the high error rate of visually identified samples, application of methods which employ material identification provide the most robust data set. Studies can optimize material identification further by applying the appropriate characterization instrument based upon the smallest particle size desired to characterize. Micro-Raman and Raman have shown interference of material identification due to biofilms or dyes influencing the monochromatic laser (Lenz et al., 2015). However, automated single-particle exploration (APSE) micro-Raman has been shown to produce robust results with more accurate quantification of MPs <500 µm (Cabernard et al., 2018). Micro-FTIR offers robust data analysis when coupled with two spectral databases (Primpke et al., 2017). Pyrolysis GC-MS is effective in identifying polymer composition; however, the destruction of samples is undesirable. Although physical stress tests may be applicable for larger particles, i.e. >1000 µm, this method is less reliable with decreasing particle size and shown in the "other" category.

With consideration for error sources, current microplastic inventory could be overestimated or underestimated. As the size range of studies approach the nanoplastic

region, more sensitive and automated methods must be applied in order to truly quantify existing plastic exposure, predict future plastic exposure, prevent human bias, and assess the hazard of environmental MNPs.

Abbreviations.

SPT, Sodium Polytungstate; PP, polypropylene; LDPE, low density polyethylene; HDPE, high density polyethylene; PS, polystyrene; Nylon, Nylon 12, 11 and Nylon 6 6,6; PMAA, poly(methyl) methacrylate; PET, polyethylene terephthalate; PVC, polyvinylchloride; PU, polyurethane; PES, polyester; PVF, polyvinyl fluoride; PTFE, polytetrafluorethylene; FTIR, Fourier Transform Infrared Spectroscopy; Pyr. GC-MS, Pyrolysis Gas Chromatography-Mass Spectrometry; EDS, Energy Dispersive X-ray Spectroscopy

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APPENDIX A

META-ANALYSIS INVENTORY

Particle. Collection Size	Min. Processing Pore Size	Min. Particle Size	Oxidative Solutions	Density Separation	I.D. Type	Ref.
0.125	0.125	0.125	30% H_2O_2 w/Fe(II) (WPO)	N/A	Visual Only (VO)	1
0.333	0.00022	ND	30% WPO	NaCl (r=undefined)	VO	2
0.125	0.125	0.125	30% WPO	ND	VO	3
0.02 (I) 0.045 (II) Bulk (III)	0.02	ND	N/A	N/A	Visual & FTIR (V&FTIR)	4
0.3	0.3	0.3	N/A	N/A	V&FTIR	5
Bulk	0.0002	0.15	30% WPO	N/A	µ-FTIR	6
0.01	0.0002	0.02	35% WPO (A) NaOH, HOCl (37%) (B)	ZnCl (r=1.6 g/cm^3) (A) NaCl (r=1.14 g/cm3) (B)	V&µ-FTIR	7
Bulk	0.075	ND	N/A	NaCl (C=120 g/L)	Other	8
0.333	0.333	0.355	N/A	ND	Other	9
Bulk	0.0012	0.1	30% WPO and 40% HF	N/A	VO	10
Bulk (I) 0.333 (II)	0.0012	< 0.5	30% WPO	ND	V&FTIR	11
Bulk	0.063	ND	N/A	NaCl (r=ND)	VO	12
2	0.00075	< 0.05	N/A	N/A	µ-FTIR	13
0.153	0.063	0.063	WPO	NaCl (r=undefined)	VO	14
Bulk	0.0625	ND	N/A	N/A	VO	15
Bulk	0.011	0.598	WPO	N/A	V&FTIR	16
Bulk (I), 0.08 (II.i) and 0.33 (II.ii)	0.0016	0.1 (I, II.i) and 0.5 (II.ii)	N/A	N/A	VO	17
0.025	0.025	0.025	WPO	NaCl (r=1.49 g/cm^3)	V&FTIR	18
Bulk	0.02	0.02	N/A	N/A	VO	19

Bulk	0.035	0.044	N/A	NaI (r=1.6 g/cm^3)	V&μ-FTIR	20
0.333	0.355	0.355	WPO	NaCl (r=1.62 g/cm^3)	VO	21
3	0.001	3	N/A	N/A	GC-MS&μ-FTIR	22
< 5	0.05	ND	N/A	ND	FTIR	23
Bulk	0.0007	<1	N/A	NaCl (C=120 g/L)	μ-FTIR	24
Bulk	1	<1	35% WPO	NaCl (r=1.2 g/cm^3)	V&GC-MS	25
Bulk	-ND	0.781	N/A	ND	μ-Raman	26
Bulk	0.032	< 0.1	N/A	(A) NaCl (r=1.2 g/cm^3) (B) Ludox-TM 40 (r=1.16 g/cm^3)	V&FTIR	27
<1 capture	0.00075	0.05	N/A	NaCl (r=2.16 g/cm^3)	FTIR	28
Bulk	-ND	ND	N/A	NaCl (r=ND)	μ-FTIR	29
Bulk	0.0016	<0.020	N/A	NaCl (C=1.18 g/L)	V&FTIR	30
Bulk	0.0012	< 0.1	30% WPO	ZnCl (r=1.5 g/cm^3)	VO	31
Bulk	ND	< 1	N/A	NaCl (r=ND)	FTIR	32
0.5	ND	< 1	N/A	N/A	VO	33
Bulk	ND	2	N/A	NaCl (r=1.2 g/cm^3)	V&FTIR	34
Bulk	0.25	< 0.25	N/A	NaCl (r=1.2 g/cm^3)	FTIR	35
2.5 x 3.5	0.001 m	0.05	N/A	NaCl (C=140 g/L)	V&FTIR	36
Bulk	ND	~4	N/A	N/A	V&FTIR	37
Bulk	ND	0.4	N/A	N/A	Other	38
1	ND	1	N/A	N/A	VO	39
ND	ND	ND	N/A	N/A	V&FTIR	40
0.333 (A) 0.25 (B)	0.2 (A) - (B)	0.25 (A&B)	N/A	N/A	V&FTIR	41

0.35	ND	>0.3	ND	ND	V&FTIR	42
<2	0.0012	ND	N/A	N/A	V&FTIR	43
0.001	N/A	0.06	N/A	N/A	μ-FTIR	44
0.35	0.1	>0.3	N/A	N/A	VO	45
Bulk	0.001	ND	N/A	NaCl (C=140 g/L)	V&μ-FTIR	46
N/A	N/A	<5	N/A	N/A	VO	47
Bulk	0.315	0.315	30% WPO	NaCl (r=1.6 g/cm^3)	FTIR	48
0.01	0.01	0.01	N/A	N/A	V&μ-Raman	49
Bulk	0.00045	ND	N/A	NaCl (r=9.043 g/cm^3)	V&Raman	50
Bulk	0.063	0.63	30% WPO, H_2SO_4	NaCl (r=1.2 g/cm^3)	V&FTIR	51
N/A	N/A	ND	N/A	N/A	FTIR	52
Bulk	1	0.1	N/A	NaCl (r=1.2 g/cm^3 and 1.8 g/cm^3)	V&GC-MS	53
Bulk	0.063	>0.063	N/A	ND	VO	54
Bulk	0.055	ND	30% WPO	$CaCl_2$ (r=undefined)	VO	55
0.355	N/A	0.41	N/A	$CalCl_2$ (r=undefined), $SrCl_2$ (r=undefined), C_2H_6O (r=undefined)	VO	56
Bulk	N/A	<1	N/A	NaCl (C=300 g/L)	V&μ-FTIR	57
Bulk (S) (W)	0.0007	ND	N/A	NaCl (r=1.2 g/cm^3)	VO	58
Bulk	0.0008	ND	30% WPO	NaCl (C=250 g/L)	VO	59
0.3 (W) 5 +- (S)	0.0016	ND	N/A	NaCl (r=1.2 g/cm^3)	VO	60
Bulk	0.025	>0.063	N/A	Sodium polytungstate (SPT) (r=undefined)	Other	61

Bulk	0.0012	ND	N/A	$ZnCl_2$ (r=1.7 g/cm^3)[f]	V&Raman	62
Bulk (S) 0.08 (W)	0.065 (S)	ND	N/A	NaCl (r=ND)	VO	63
N/A (A) Bulk (B)	N/A (A) (B)	<10 (A) <0.05 (B)	N/A (A) (B)	N/A (A) SPT (r=1.5 g/cm3) (B)	V&Raman& FTIR	64
Bulk (S) 0.333 m (W, A) bulk (W, B)	0.1 m (W, A) 0.005 (W, B) 0.005 (S)	0.333 (W, A) 0.005 (W, B) (S)	30% WPO (W, A) (W, B) (S)	NaCl (r=ND) *C=360 g/L*	V&μ-FTIR&SEM-EDS	65
Bulk	0.0012	<0.5	N/A	$KHCO_2$ (r=1.5 g/cm^3)	V&Raman	66
0.3	0.3	ND	Novel method [c]	NaCl (r=1.16 g/cm^3)	V&FTIR	67
Bulk	0.063	ND	N/A	NaCl (r=ND)	VO	68
Bulk (All)	0.0002 (All)	<0.3	N/A	NaCl (r=1.2 g/cm^3)	V&FTIR	69
0.25	0.0012	<1.25	N/A	N/A	V&Raman	70
Bulk	5	0.1	30% WPO	NaCl (r=1.3 g/cm3)	μ-Raman	71
Bulk	0.174	ND	30% WPO	ZnCl (r=1.6 g/cm3)	VO	72
Individual extraction	ND	ND	ND	ND	V&FTIR	73
Bulk	0.038	0.038	N/A	NaCl (r=ND)	V&FTIR	74
0.3	N/A	0.3	30% WPO	NaCl (C=300 g/L)	V&Raman	75
0.333	0.0007	0.5	N/A	N/A	V&μ-Raman	76
Bulk	0.001	ND	N/A	NaCl (r=ND)	V&μ-FTIR&SEM-EDS	77
Bulk	0.001	0.1	N/A	NaCl (r=1.27 g/cm3)	V&FTIR	78
Bulk	0.25 m	0.25	N/A	NaCl (r=1.2 g/cm3)	VO	79

Bulk	0.00045	<1	N/A	NaCl (r=1.2 g/cm3)	GC-MS&SEM-EDS	80
Bulk (S) 0.3 (W)	0.3	>0.3	35% WPO	NaCl (r=ND)	V&GC-MS&FTIR	81
0.2	N/A	0.3	N/A	N/A	V&Raman	82
Bulk (A) 0.05 (B) 0.33 (C) 2 (D)	0.00075	ND	34.5% WPO	N/A	V&μ-FTIR	83
0.35	0.3	0.3	N/A	N/A	V&FTIR	84
2 (A) 0.02 (B) 0.33 (C)	0.00075	0.05	30% WPO (B) (C)	N/A	V&FTIR	85
0.05	0.045	0.05	30% WPO	N/A	V&FTIR	86
0.3	0.3	0.3	N/A	NaCl (r=ND)	VO	87
0.333	0.0007	0.33	30% WPO	NaCl (C=6 mol/L)	V&GC-MS	88
0.33 (A) 0.05 (B)	0.0007	2 (A) 0.05 (B)	20% WPO	N/A	V&FTIR	89
0.3	ND	0.1	N/A	ND	V&FTIR	90
0.333	N/A	0.333	N/A	N/A	VO	91
Bulk	0.02	0.02	30% VIP1[h]	ND	VO	92
Core	0.00022	<0.2	N/A	N/A	V&FTIR	93
0.25	0.0012	<1.25	N/A	N/A	V&Raman	94
0.33	N/A	0.32	N/A	NaCl (r=ND)	VO	95
Bulk	0.174	0.5	N/A	N/A	VO	96
Bulk	0.174	0.5	30% WPO	ZnCl (r=1.6 g/cm3)	VO	97
Bulk	0.00045	0.01	N/A	NaCl (r=1.2 g/cm3) NaI (r=1.8 g/cm3)	V&FTIR	98
Bulk	0.001	ND	30% WPO	NaCl (r=ND)	μ-FTIR	99
0.112 (W) 0.3 (S)	0.0012 (W) (S)	0.112	N/A	$KHCO_2$ (r=1.5 g/cm^3)	V&Raman	100
Bulk	0.01	≤ 0.011	30% WPO	ZnCl (r=1.7-1.8 g/cm3)	V&μ-FTIR&ATR-FTIR	101

Bulk	0.063	0.25	N/A	SPT (r=ND)	V&FTIR	102
Bulk	0.002	≤ 0.1	N/A	NaCl (r=1.2 g/cm3) NaI (r=1.6 g/cm3)	SEM-EDS	103
Bulk	0.5	ND	N/A	ND	VO	104
Bulk	ND	0.3	N/A	N/A	V&FTIR	105
Bulk	0.0012	0.063	30% WPO	NaI (r=1.6-1.8 g/cm3)	VO	106
Bulk	0.0007	ND	N/A	NaCl (r=ND)	V&FTIR	107
0.1 (W), Bulk (S)	0.03 (W), 0.1 (S)	0.1 (W), <0.005 (S)	30% WPO (W) (S)	NaCl (r=undefined)	VO	108
Bulk	0.00045	0.06	N/A	NaCl (r=1.2 g/cm3) NaI (r=1.8 g/cm3)	V&FTIR	109
Bulk	0.007	0.51	N/A	NaCl (r=ND)	VO	110
0.3 (W) Bulk (S)	0.02	£ 0.5	N/A	NaCl (r=ND)	V&FTIR	111
Bulk	0.01	0.02	N/A	NaCl (r=ND)	VO	112
Bulk	0.063	<0.063	N/A	NaCl (r=ND)	Other	113
Bulk	0.063	0.063	N/A	NaCl (r=ND)	VO	114
0.15	0.02	0.1	30% KOH and NaClO	N/A	Other	115
0.1	ND	1	N/A	N/A	V&FTIR	116
0.333	0.25	0.333	30% WPO	N/A	V&SEM-EDS	117
0.01	0.01	ND	sodium dodecylsulfate solution	N/A	µ-Raman	118
0.315	N/A	ND	N/A	N/A	VO	119
0.3	0.0045	0.5	N/A	N/A	V&FTIR	120
0.5	N/A	ND	N/A	ND	VO	121
individual extraction	1	2	N/A	N/A	V&FTIR	122
0.333	0.0007	0.24	N/A	N/A	V&FTIR	123
Bulk	0.47	ND	N/A	NaCl	VO	124

0.33	0.0007	0.1	30% WPO	N/A	V&FTIR	125
3	0.5	0.5	N/A	N/A	V&Raman	126
Bulk	0.032	1	N/A	NaCl	V&μ-Raman	127
0.125	0.0008	ND	30% WPO	N/A	V&μ-FTIR	128
Bulk	0.0002	ND	N/A	Sewater	V&FTIR	129
Bulk	0.005	ND	5% HCl	NaCl (r=1.17g/cm3)	V&FTIR	130
Sweeping	0.002	0.05	30% H2O2	ZnCl2(c=1.78 kg/L)	V&SEM-EDS	131
ND	ND	ND	N/A	N/A	FTIR	132
Bulk	0.00045	0.02	H2O2	NaCl (c=1.18 g/l)	VO	133
Core	0.0012	ND	30% WPO	NaCl (r=1.6-1.8 g/ml)	Other	134

APPENDIX B

VISUAL IDENTIFICATION ERROR ESTIMATION

Error	(%)	*n*=particle count	Over or underestimate	Reference
Misidentification <50 micrometer	37.0	637	Overestimate	118
Misidentification 50-100 micrometer	33.0	155	Overestimate	118
Misidentification >100 micrometer	17.0	35	Overestimate	118
Degradation or misidentification	82.5	177	Overestimate	101
Misidentification	20.0	20	Overestimate	19
Misidentification	47.0	32	Overestimate	53
Fragment error SML	38.3	206	Underestimate	131
Fragment error beach	87.2	1192	Underestimate	131
Fiber SML	65.4	13	Overestimate	131
Fiber beach	72.4	29	Overestimate	131
Misidentified as plastic	39.0	674	Overestimate	129
Microplastics identified as ash, GL21	27.0	57	Overestimate	9
Microplastics identified as ash, GL20	28.0	1101	Overestimate	9
Microplastics identified as ash, GL19	11.0	21	Overestimate	9
Microplastics identified as ash, GL14	31.0	26	Overestimate	9
Microplastics identified as ash, GL10	18.0	15	Overestimate	9
Microplastics identified as ash, GL9	25.0	5	Overestimate	9
Microplastics identified as ash, GL7	4.0	3	Overestimate	9

APPENDIX C

DENSITY SEPARATION MISMATCH

Sample Environment	Solute Density (g/cm^3)	Ref.
Water	1.14	7
Water	1.16	67
Water	1.2	69
Sediment	1.2	25
Sediment	1.2	35
Sediment	1.2	58
Sediment	1.3	71
Sediment	1.27	78
Sediment	1.2	79
Sediment	1.2	80
Sediment	1.2	98
Sediment	1.2	109
Sediment	1.17	130

APPENDIX D

MATERIAL AND VISUAL CHARACTERIZATION SIZE LIMITATIONS

Characterization Type	Instrument	Minimum Effective Size	Limitations
Spectroscopy	ATR-Fourier Transform Infrared Spectroscopy (FTIR)	1 mm	-
Spectroscopy	Raman Spectroscopy	1 mm	-
Micro-Spectroscopy	Microscope attached to FTIR	15 mm	-
Micro-Spectroscopy	Microscope attached to Raman	0.5 mm	Pigmentation causes interference
Spectroscopy-Microscopy	Scanning Electron Microscope with Energy Dispersive X-ray Spectroscopy (SEM-EDS)	0.001 mm	Possibly destructive
Microscopy	Atomic Force Microscope (AFM)	0.01 mm	Only exterior interrogation
Microscopy	Light Microscope	0.5 mm	Only exterior interrogation
Microscopy	Stereo or Dissecting Microscope	1 mm	Only exterior interrogation
Spectrometry	Pyrolysis GC-MS	0.001 mm	Physically placing particle with tweezers Destructive