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Simplifying Microplastic via Continuous Probability Distributions for Size, Shape, and Density

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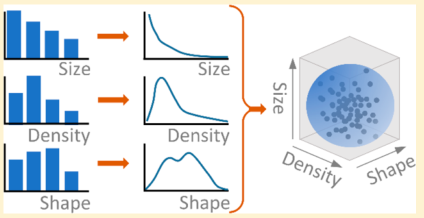
Supporting Information

ABSTRACT: Because of their diverse sizes, shapes, and densities, environmental microplastics are often perceived as complex. Many studies struggle with this complexity and either address only a part of this diversity or present data using discrete classifications for sizes, shapes, and densities. We argue that such classifications will never be fully satisfactory, as any definition using classes does not capture the essentially continuous nature of environmental microplastic. Therefore, we propose to simplify microplastics by fully defining them through a three-dimensional (3D) probability distribution, with size, shape, and density as dimensions. In addition to introducing the concept, we parametrize these probability distributions, using empirical data. This parametrization results in an approximate yet realistic representation of "true" environmental microplastic. This approach to simplifying microplastic could be applicable to exposure measurements, effect studies, and fate modeling. Furthermore, it allows for easy comparison between studies, irrespective of sampling or laboratory setup. We demonstrate how the 3D probability distribution of environmental versus ingested microplastic can be helpful in understanding the bioavailability of and exposure to microplastic. We argue that the concept of simplified microplastic will also be helpful in probabilistic risk modeling, which would greatly enhance our understanding of the risk that microplastics pose to the environment.



INTRODUCTION

Microplastic particles are complex and diverse.[1] Their size, shape, and density vary along continuous scales. Additionally, microplastics consist of mixtures of polymers and chemical additives at various states of weathering. When microplastics are exposed in the natural environment, absorption of chemical contaminants and the formation of biofilms further enhance their complexity.[2,3] Consequently, the characteristics of environmental microplastics can be considered to cover a continuous parameter space rather than a limited set of discrete values. The research community sometimes struggles with the definition of microplastics.[4] Several recent reviews characterize microplastic particles using discrete size fractions, polymer types, and shape characteristics.[3,4] Such classifications using discrete categories may be useful for describing approximate properties or in regulatory frameworks.[5] However, we argue that such characterizations will never be fully satisfactory. After all, any definition using classes does not capture the continuous nature of microplastics. Whatever boundaries are taken, within those boundaries the actual distribution still can be manifold. This is perhaps most conspicuous for particle size, but microplastic density and shape also have a continuous nature. Defining a polymer type may be useful when referring to pristine materials or studying the origin of particles but is less useful when studying the physical properties of environmental microplastic. After all, in the environment, each original polymer has become more or less dense due to weathering, embrittlement, and biofouling.[6] Shape is continuous, as well, and no classification (e.g., beads, fibers, foam, fragments, and sheets) will ever be able to accurately capture the full variability of shapes.

Here, we propose that environmental microplastics can best be simplified using a set of distribution functions capturing their main characteristics along continuous scales. This concerns the characteristics of the particles and not the abundance in the environment. The abundance of microplastic is highly relevant to understand the risks of the presence of the particles; however, this is not as such a characteristic of the material, and microplastic abundance has been studied and summarized extensively previously.[3,7,8] The aim of this paper is to demonstrate the added value of simplifying microplastics using continuous distributions for size, density, and shape. In addition to introducing the concept of environmental microplastic via probability distributions, we propose mathematical functions that can capture the measured variability of microplastics with respect to size, shape, and density. Distribution functions are selected and fitted, which represent empirical data on microplastics more accurately than when using discrete categorizations. This leads to a characterization that is more loyal to the real material. We discuss how the concept may be used in fate modeling, exposure and effect assessments, and, ultimately, risk assessment.

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MATERIALS AND METHODS

Simplifying Size. Particle size distributions of natural particles in aquatic systems have been studied extensively. Different distributions have been found, with log-normal, bimodal, and power law distributions proven to be effective models.[9−11] Microplastic is usually expected to also show a power law distribution, because fragmentation causes the particles to break down in ever smaller pieces.[12−14] Hence, we fitted a power law curve to data of peer-reviewed studies. These studies were found using databases such as Scopus and Google Scholar, with the final search in May 2019. For a study to be included, results had to be presented in ≥10 size bins in the microplastic size range (<5 mm) to allow for meaningful data fitting. The exponent α was fitted using

$$\log(y) = \log(b) - \alpha \log(x), \text{ which equates to } y = bx^{-\alpha} \tag{1}$$

where x is the particle size (micrometers) in its longest dimension and y is the abundance (percent). Fitted values for b were not very relevant, because they depend on the unit of y (e.g., abundance in percent, number, or number per cubic meter). However, b could be rewritten as a function of $x_{\min}$ and α:

$$b = (\alpha - 1){x_{\min}}^{\alpha-1} \tag{2}$$

where $x_{\min}$ is the minimum particle size for which the equation is valid. Linear regression was used to assess the values and statistical significance of α.

Simplifying Shape. Microplastics are often classified in several shape categories, commonly named fragments, fibers, films, foam, and beads.[3] However, no universal definition exists to classify particles on the basis of shape, whereas per category the actual shapes still are highly diverse.[4,15] Additionally, while they might be useful for legislation, descriptive shape categories have little meaning when studying the fate of particles. Therefore, we propose a more generalized approach using length:width:height (L:W:H) ratios for the different shape categories. Being the largest dimension, all lengths were assigned a value of 1, resulting in widths and heights of ≤1. Because the relative dimensions of each shape category vary, we included upper and lower limits for the particle widths and heights. For fragments and foam, a W:H ratio of 1 was assumed as an upper limit, while for the lower limit, we assumed the W:H ratio to be equal to the L:W ratio.[16] Fibers are usually cylindrical with respect to their primary shape (i.e., not coiled up), so the W:H ratio = 1. The L:W = H ratio can vary between 0.5 and 0.001.[17−19] Films were assumed to have an L:W ratio similar to that of fragments but usually are thinner. Therefore, the height was assumed to be 10 times smaller than that for fragments. This assumption is valid for microplastics, because a 5 mm film with a thickness of 20 μm, a thin plastic bag, results in an L:H ratio of 0.004, which falls inside our range. Finally, beads vary between perfect spheres with an L:W:H of 1 to more ellipsoidal particles. On the basis of analysis of published pictures with ImageJ, an L:W ratio of 0.60 was found for beads.[20] Again assuming a similar ratio for the W:H ratio, the smaller height limit was set at 0.36. Please note that these ratios were defined for microplastic, not macroplastic. A plastic bag (30 cm) can still be 20 μm thin and would thus have a ratio of 10^{-5}. For these larger plastics, it is better to use the actual size instead of normalized ratios.

We assumed a triangular distribution to capture the variability in each shape category, with the upper and lower limits as the minimum and maximum, respectively, and the most abundant shape in the middle of the lower and upper limit.[21,22] On the basis of the resulting L:W:H ratio distributions, we calculated dimensionless Corey shape factor (CSF) distributions:[23,24]

$$\text{CSF} = \frac{H}{\sqrt{LW}} \tag{3}$$

Other shape factors have also been applied to microplastics, such as the aspect ratio,[25] the equivalent spherical diameter,[26] or a shape factor based on the surface area and perimeter.[27] By using the CSF, some of the key implications of shape for fate processes are preserved, such as the three-dimensional (3D) aspect needed to describe settling behavior. For the relative abundances of shape categories, their average abundances in water (based on 46 studies) and sediment (based on 45 studies) were used.[3] Given this high number and the diversity of the studies used, the data cover a large range of locations as well as a wide range of ages and weathering stages of the plastic particles.[28,29] By combining the triangular shape factor distributions and the relative abundance of each of the shape categories, we generated a universal shape distribution for environmental microplastic (random number generator; $n = 10^7$). The distribution showed a bimodal shape (eq 4), which was fitted using the package mixtools in RStudio.[30,31]

$$y = f_1 \frac{1}{\sqrt{2\pi{\sigma_1}^2}} e^{-(x-\mu_1)^2/2{\sigma_1}^2} + f_2 \frac{1}{\sqrt{2\pi{\sigma_2}^2}} e^{-(x-\mu_2)^2/2{\sigma_2}^2} \tag{4}$$

where f, μ, and σ are the relative contribution, mean, and standard deviation of the two normal distributions, respectively. A Pearson χ^2 test was used to test if the fit of the modeled distribution to the data was statistically significant.

Simplifying Density. Data on the abundance of microplastic polymer types in water and sediment were combined with lower and upper density limits of pristine polymers, as reported in the literature. The effect of weathering, e.g., biofouling, ultraviolet degradation, or mechanical abrasion, on polymer density was taken into account using the work of Kaiser et al.[32] According to their work, the settling velocity of polystyrene particles (PS, 1 mm) can increase by 81% when they are fouled. This corresponds to an 81% increase in the density difference between the particle and water, assuming a constant size and flow regime. We applied this percent increase to the upper density limit of all nonfloating polymers. Floating polyethylene particles (PE, 1 mm) were found to settle after fouling, with settling velocities as high as 0.04 m/s.[32] Given this settling velocity, the water density, and the particle size, we estimated an equivalent particle density of 1.099 g/cm^3 based on the Stokes settling equation. Because the initial particles had a density of 0.955 g/cm^3, the density increased by 15%. We applied this density increase percentage to the upper limit of all floating polymers.

All common polymer types have been found in surface water samples.[3,33] This indicates that despite the lower limit of pristine polymer density exceeding that of water, they remain in suspension. Because very little is known about the degradation and the subsequent density decrease of polymers, we assumed a lower limit of 1 g/cm^3 for these polymers (summarized in Table S3). Triangular distributions for the

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density of individual polymers were assumed, with the upper and lower limit as the minimum and maximum, respectively, for the distribution. Random data points ($n = 10^7$) were generated with the triangular distributions and the relative abundance of each polymer type as inputs.

The resulting continuous density distribution showed a normal-inverse Gaussian shape, which was fitted accordingly (eq 5).

$$y = \frac{\alpha\delta K_1\left[\alpha\sqrt{\delta^2 + (x-\mu)^2}\right]}{\pi\sqrt{\delta^2 + (x-\mu)^2}} e^{\delta\sqrt{\alpha^2-\beta^2}+\beta(x-\mu)} \tag{5}$$

where μ, δ, α, and β represent the location, scale, tail heaviness, and asymmetry of the distribution, respectively. K_1 is a modified Bessel function of the third kind with order 1.[34] A Pearson χ^2 test was used to test if the fit of the modeled distribution to the data was statistically significant.

RESULTS AND DISCUSSION

A Probability Distribution for Size. Nineteen particle size distributions with data for ≥10 size bins were available from 11 studies, with some studies presenting their results separately for different locations or plastic types (Table S1).[35−45] The data of these studies showed one of two different patterns. Either they showed a decrease in particle concentration with an increase in size, or they found an initial increase in concentration with particle size, followed by a decrease similar to the first mentioned pattern. There are two explanations for this initial increase. First, small microplastics at the surface of the water are more susceptible to wind mixing, fouling, and aggregation,[46−48] which would decrease their abundance. Second, the smallest particles are easily overlooked in the sample analyses, leading to an analysis bias, especially because some studies used visual inspection for <0.5 mm particles.[49] However, because we aim to address environmental microplastic particles, and not just those at the water surface, and because underestimation of abundances in the lower size ranges is likely, we here included only the data showing a decreasing concentration with an increasing particle size. Because number concentrations are lower for larger particles (Figure 1), in some studies the detection limit was reached for these larger particles. Below this limit, detecting a single particle is a matter of chance, leading to binary number concentrations (i.e., irregularly intermittent 0 and 1 counts), which can easily be seen in the data.[40−42] We calculated the detection limit of these studies and included only data higher than the detection limit (explanation provided as Supporting Information). The fitted power law resulted in an average ± standard deviation exponent $\alpha = 1.6 \pm 0.5$ ($n = 19$) (Figure 1). The goodness of fit (R^2) varied between 0.53 and 0.97 (Table S4). For microplastic, we propose that x_{min} (eq 2) equals 20 μm, which is a common detection limit for the analysis of microplastics using Fourier transform infrared spectroscopy.[50,51] Because the function has an infinite tail toward the larger particles, a cutoff at 5 mm was assumed. Theoretically, the slope (α) can range between 0 and 3, with a value of 3 representing 3D fragmentation with full conservation of mass.[14,52] Therefore, a value of 1.6 suggests that fragmentation of plastic might partly be two-dimensional (2D) (e.g., for sheets).[52] Additionally, the size distribution not only is driven by fragmentation but also depends on size selective emissions and processes.[52] The agreement in the value of α across data sets confirms our hypothesis with respect to the generic features of the microplastic size distributions, which subsequently can be used to generalize this aspect of the material for prospective purposes. With lower and upper boundaries of 20 μm and 5 mm, respectively, this results in a generic continuous particle size distribution, which can be compared to any study reporting continuous data, independent of the sampling design.

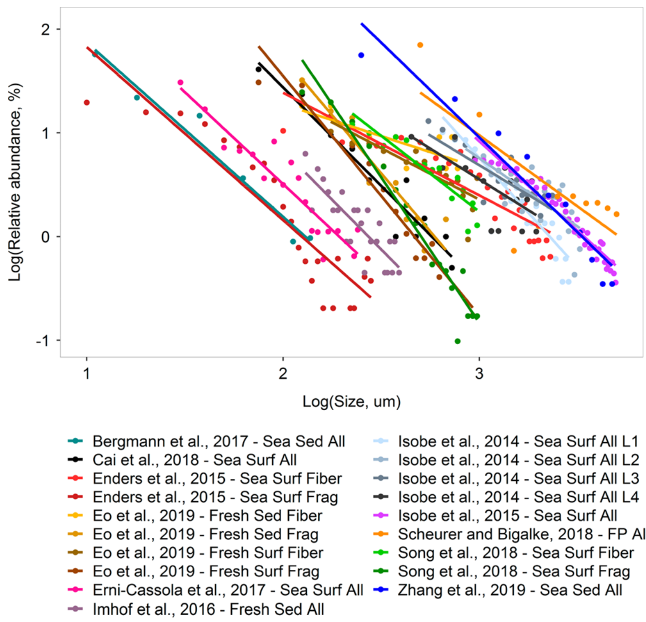

Figure 1. Relative microplastic abundance for different particle sizes. The fitted trend lines, $y = bx^{-\alpha}$ or $\log(y) = c - \alpha \log(x)$, have similar slopes on a log−log scale, with an average exponent value of $\alpha = 1.6$ ($n = 19$). The legend refers to the marine ("Sea") or freshwater ("Fresh") environment, where samples were taken in the water ("Surf") or in the sediment ("Sed"). Some studies differentiated between different plastic types, namely, fibers ("Fiber") and fragments ("Frag"). Studies that did not specify their plastic types are labeled "All". FP refers to floodplain. L1−L4 refer to four different locations studied by Isobe et al.[42] The data and the data selection steps are available in the Supporting Information. A graph including all data points, that is, including zero values and data below and beyond the detection limits, is included in the Supporting Information (Figure S1).

A Probability Distribution for Shape. The most abundant shape category of microplastic in water and sediment is fibers (48.5%), followed by fragments (31%), beads (6.5%), films (5.5%), and foam (3.5%).[3] Combining these abundance data with the triangular shape distributions (Table S2 and Figure S2) resulted in a continuous bimodal microplastic shape distribution (Figure 2A). The fitted parameter values for eq 4 were as follows: $f_1 = 0.06$, $f_2 = 0.94$, $\sigma_1 = 0.03$, $\sigma_2 = 0.19$, $\mu_1 = 0.08$, and $\mu_2 = 0.44$ (see Table S5 for standard errors). A Pearson χ^2 test indicated that the optimized distribution fits the data well (the fitted model did not differ significantly from the data; $p = 0.231 > 0.05$). The distribution is dominated by fibers and fragments (CSF = 0.25−0.75) but also has a distinct second peak at a CSF of 0.07, which is mainly attributed to sheets (Figure 2A). The distribution captures the main features of shapes encountered in the environment as well as the relative abundances of these (now continuous) shapes in one go. Most illustrative though is the continuous character of microplastic shape.

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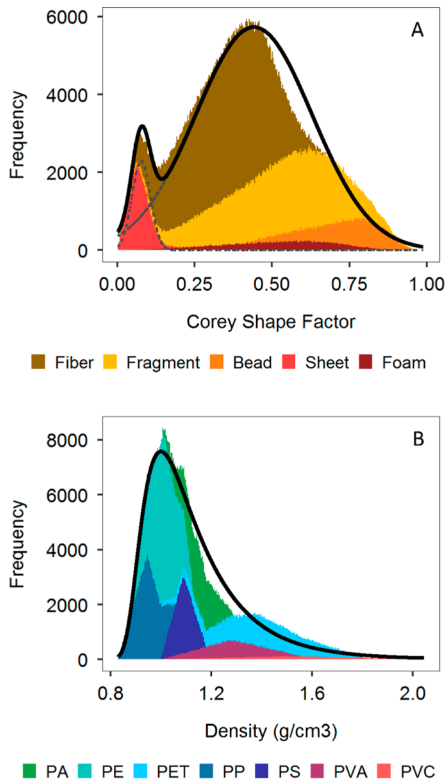

Figure 2. Distributions and fitted models for randomly generated data ($n = 10^6$). (A) Shape distribution of microplastic, expressed as Corey shape factor. A bimodal distribution was fitted through the data (solid line), which is the sum of two normal distributions (dotted lines). (B) Density distribution of microplastic, with a fitted normal-inverse Gaussian distribution (solid line).

A Probability Distribution for Density. The most common microplastic polymer types in the aquatic environment are PE (25%), PET (16.5%), PA (12%), PP (14%), PS (8.5%), PVA (6%), and PVC (2%).[3,33] Lower and upper density limits (grams per cubic centimeter) of these microplastic polymer types as reported in microplastic literature are 0.89−0.98 for PE, 0.96−1.45 for PET, 1.02−1.16 for PA, 0.83−0.92 for PP, 1.04−1.10 for PS, 1.19−1.31 for PVA, and 1.10−1.58 for PVC.[6,53−56] We assumed triangular density distributions for the different polymer types, given these lower and upper limits. These distributions were combined with the occurrence percentages, which resulted in a trimodal distribution, with a clear gap around 1.0 g/cm^3 (Figure S3a). When the aforementioned effect of biofouling was included, a more continuous distribution was obtained (Figure S3b and Table S3). Finally, assuming all polymers can have a density as low as 1 g/cm^3, we obtain a continuous density distribution (Figure 2B). The normal-inverse Gaussian distribution was fitted, resulting in the following parameter values for eq 5: $\mu = 0.84$, $\delta = 0.097$, $\alpha = 75.1$, and $\beta = 71.3$ (see Table S6 for standard errors). The normal-inverse Gaussian distribution with the parameters mentioned above significantly fitted the data, because the Pearson χ^2 test gives $p = 0.234 > 0.05$, so the data did not differ significantly from the model. The distribution successfully captures the continuous nature of microplastic densities in the environment.

Environmental Microplastic as a Three-Dimensional Probability Distribution of Size, Shape, and Density. With these defined distributions for microplastic size, shape, and density, we can illustrate the concept of microplastic as a probability distribution in three dimensions, a 3D microplastic space. Independent random numbers ($n = 10^4$) were generated for microplastic size, shape, and density, using the newly obtained distributions as input. Here, we used the best estimate for the parameter values, but the standard errors can also be utilized to further account for the uncertainty and diversity in these estimates. We assumed size, shape, and density to be independent, because a large majority of plastic is secondary and all particles are subject to similar weathering processes. The 3D probability distribution was created by plotting each combination of the three plastic properties (Figure 3A). Additionally, 2D kernel density contour plots

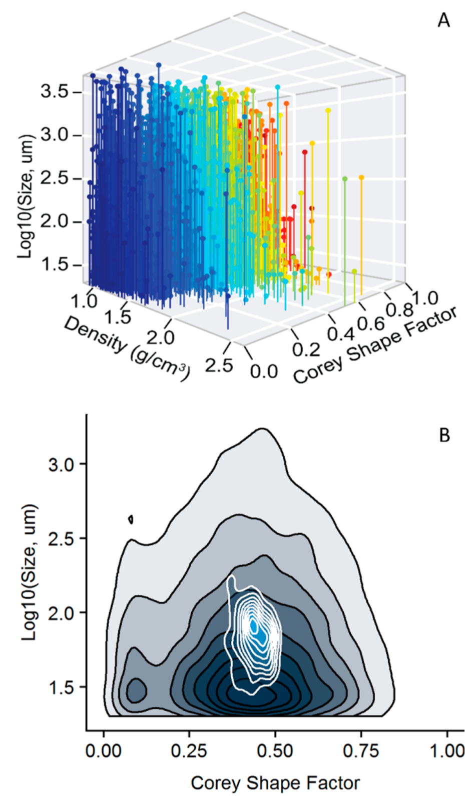

Figure 3. (A) Environmental microplastic as a probability distribution in three dimensions, defining the features of microplastic in the 3D parameter space. Colors indicate the CSF from 0 (blue) to 1 (red). The dots are the randomly generated points ($n = 10^4$) in the 3D space, and the lines illustrate the location on the $x-y$ plane. (B) Kernel density plot of the same parameter space, with the black contours representing the kernel density for the whole microplastic space and the white contours for those of only the microplastics found in the body of *Gammarus pulex*.[57]

were obtained for each combination of the plastic properties (Figure 3B and Figure S3). In the contour plots, the imposed boundaries for particle size (20−5000 μm) and shape (0−1) are visible. On average, environmental microplastic appears to have a density of ∼1 g/cm^3, a size of 20 μm, and a CSF of 0.4 (Figure 3 and Figure S4).

Merits and Limitations of Microplastic Represented as 3D Probability Distributions. Fate modeling, exposure assessments, effect studies, and risk assessments may benefit from representing environmental microplastic as a distribution in 3D parameter space. Recognizing the 3D microplastic space as depicted in Figure 3A may be helpful in communicating the

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continuous nature of microplastic, which makes the material difficult to understand and different from other environmental stressors. At the same time, the concept we propose is a substantial simplification of the current approaches. After all, the number of parameters required by the three mathematical functions (n = 12 parameters) is far lower than the number of descriptors currently used to define the numerous discrete shape, size, and polymer identities of environmental microplastic. Furthermore, this replacement by continuous functions has an advantage in that it interpolates between discrete categorical descriptors and thus is more accurate.

Studies of microplastic abundance and fate can report data as distributions and compare them with the here reported parametrized distribution functions. For example, a recent study illustrated the use of continuous distributions for microplastic settling velocities.[58] Another example is that effect studies often consider the size fraction that is bioavailable[27,57] and the effect of particle morphology on ingestion and egestion rates.[59,60] Such size fractions or shapes now can easily be selected from the generic distributions for different species, such that they best fit their species-specific traits. For instance, we can plot the characteristics of the microplastic ingested by *Gammarus pulex*, as found by Redondo-Hasselerharm et al., in a 2D kernel density plot (Figure 3B). From this figure, we can see that a large portion of the size range of environmental microplastic is bioavailable for *Gammarus* and that the organisms mainly ingested the particles of the most common shape (Figure 3B).[57] With this kind of comparison, a clear view of which part of the parameter space would be relevant for assessing risks of microplastic exposure for this species is provided.

Model studies often use distributions to capture the variability and uncertainty of certain parameters.[61,62] We propose that the distributions presented here can be used as a first approximation of environmental microplastic, in such model simulations. We emphasize that these calibrations are provisional and probably tend toward the aquatic environment. We did not provide distributions for some other processes or properties, e.g., for complex aggregate characteristics, color, biofilm features, or chemical composition. If more data become available, parameter values may be reset or tuned to other specific environments. Also, these distributions will not necessarily be equal to microplastic distributions for all specific locations, because we aimed to represent the average material and therefore combined data sets. For many applications, including prospective modeling and risk assessments, such average characteristics are of interest. To date, probabilistic approaches have not been applied in microplastic risk research. It is, however, common practice in comparable research fields, such as for the assessment of nanoparticles[63−65] and chemicals.[66,67] The limited number of microplastic risk assessments performed so far all had to deal with limited, fragmented, and often incomparable data.[3,68,69] When future studies would strive at reporting microplastic characteristics along continuous scales, more rigorous probabilistic risk assessments could be made, which would greatly enhance the understanding of the risk that microplastic poses.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.estlett.9b00379.

Description of the data selection for the size distribution; details of the studies used for the size distributions (Table S1); size distributions including zero values and concentrations below and beyond the detection limits (Figure S1); L:W:H ratios for different shape classes (Figure S2 and Table S2); abundance and density distributions for pristine and weathered plastics (Figure S3 and Table S3); kernel density plots for microplastic size, shape, and density (Figure S4); regression statistics for the size distributions (Table S4); statistics for the bimodal distribution fitting (Table S5); and statistics for the inverse Gaussian distribution fitting (Table S6) (PDF)

All data collected from the original published figures (XLSX)

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Notes

The authors declare no competing financial interest.

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