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Advancing Evaluation of Microplastics Thresholds to Inform Water Treatment Needs and Risks

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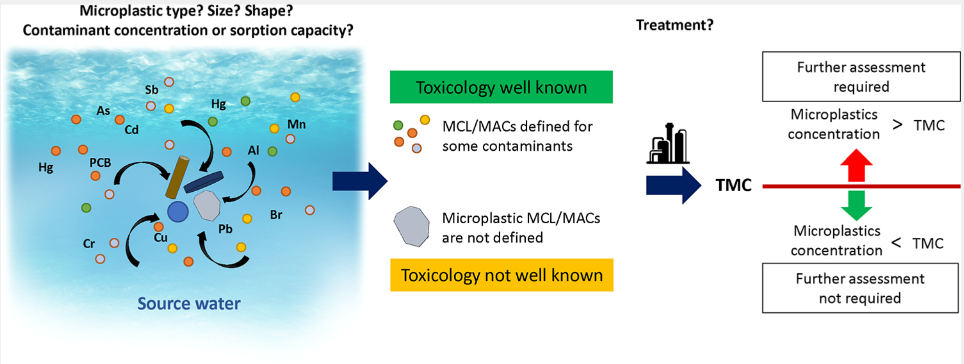

ABSTRACT: Although human health impacts of microplastics are not well understood, concern regarding chemical contaminants retained on or within them is growing. Drinking water providers are increasingly asked about these risks, but strategies for evaluating them and the extent of treatment needed to manage them are currently lacking. Microplastics can potentially induce health effects if the concentration of contaminants adsorbed to them exceeds predetermined drinking water guidelines (e.g., Maximum Contaminant Levels). The risk posed by microplastics due to adsorbed contaminants is difficult to determine, but a worst-case scenario can be evaluated by using adsorption capacity. Here, a "Threshold Microplastics Concentration" (TMC) framework is developed to evaluate whether waterborne microplastic concentrations can potentially result in the intake of regulated contaminants on/in microplastics at levels of human health concern and identify treatment targets for managing associated health risk. Exceeding the TMC does not indicate an immediate health risk; it informs the need for detailed risk assessment or further treatment evaluation to ensure particle removal targets are achieved. Thus, the TMC concept and framework provide an updateable, science-based screening tool to determine if there is a need for detailed risk assessment or treatment modification due to waterborne microplastics in supplies used for potable water production.

KEYWORDS: *drinking water treatment, chemical contaminants, adsorption capacity, risk management, maximum contaminant level*

1. INTRODUCTION

In recent years, microplastics and nanoplastics have emerged as contaminants of concern due to their ubiquitous presence, including in aquatic environments.[1] Microplastics research initially focused on marine systems; impacts of microplastics contamination on marine aquatic organisms have been documented.[2−4] This, in addition to increasing reports of microplastics presence in freshwater and drinking water,[5,6] has led to questions about potential human health risks resulting from microplastics ingestion.[7,8]

Even though humans can be exposed to microplastics through multiple routes including air, food, and beverages,[9−11] adverse effects on human health have not been conclusively reported. This is due to a lack of reliable toxicological data,[12,13] uncertainties in human exposure to microplastics,[14,15] and challenges in sampling and quantification.[16,17] Therefore, risk assessments are not currently able to assess health risk accurately.[18] Furthermore, microplastics may induce health effects in different ways: (1) the effect of the microplastic particles themselves,[19−21] (2) the effect of chemicals associated with microplastics, including adsorbed/absorbed contaminants or additives,[22−24] and (3) the potential effect of attached microbial biofilm on microplastics themselves or its role in breaking down and altering the chemical makeup of microplastics.[25−28] This highlights the necessary multidimensionality of microplastics dose−response assessment to determine their overall toxic effects. Additional complexity arises because the

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many possible combinations of chemicals and physicochemical properties make it impossible to account for all types and sizes of plastics and their association with all regulated contaminants.[29] Accordingly, a comprehensive health risk assessment of microplastics has not been conducted to date.

Due to the limitations and complexity of determining microplastic health effects, the derivation of a comprehensive health-based threshold and regulatory guideline for microplastics in drinking water is not possible at present. Despite attempts, the unavailability of reliable data such as microplastics concentration and characteristics, the extent to which humans are exposed to them, and their health implications on humans limit the scope of risk assessments and undermine confidence in enforcing regulations.[30] Most existing toxicity assessments have investigated the physical effects of microplastic particles. Physical and chemical effects of microplastics are not analyzed separately, and the results obtained from studies represent their overall toxicities.[31] Therefore, dose−response assessments are unable to identify the driver(s) of toxicity: the physical effect of microplastics themselves or the chemicals associated with them. Studies have alternately focused on creating frameworks and providing guidance for risk assessment that may be utilized once reliable data become available.[32,33] A more holistic and systematic framework to incorporate the different ways in which microplastics can have health implications[34] is necessary to understand their effects in isolation and to identify characteristics that most pose a hazard.[29] While it is not possible to fully assess health risk at present, it is possible to establish a framework for doing so and use it and available data (e.g., contaminant adsorption) to start to determine when microplastics do *not* pose a risk to drinking water.

Several contaminants have the potential to adsorb to microplastics,[35−38] and others are used as additives (including phthalates, organophosphate esters, bisphenols, and organotins) that are known to be endocrine disruptors.[39−42] These additives are not typically chemically bound to the plastic polymer; thus, their water-extractable fractions may be released to the water column. This process is largely dictated by polymer−water partitioning.[42,43] While it has been suggested that the effect of microplastics as vectors for chemical contaminants is not concerning for human health,[44] adsorbed chemical contaminants can desorb from microplastics to both the water column and the human gut—also it has been shown that contaminant desorption from microplastics can be enhanced in simulated gut environments.[45−47] Thus, microplastics ingestion may result in human exposure to other contaminants[48,49] (i.e., microplastics may act as a vector for contaminants) and induce health effects if released chemical concentrations exceed established health thresholds.[50] To date, however, most microplastics experiments focused on adsorption/desorption have not reflected natural conditions[13] nor been based on microplastics concentrations currently detected in the environment. Despite this limitation, the potential role of waterborne microplastics in contributing to chemical contaminant exposure can be explored by using data that are currently available.

While determining health-based regulatory threshold concentrations for microplastics in drinking water is at best a work in progress, a wide range of contaminants that are known to be detrimental to human health are regulated. Their Maximum Contaminant Levels (MCLs[51]) or Maximum Acceptable Concentrations (MACs[52]), among other health guidelines, have been established to limit potential health risks from drinking water and to require monitoring of source and treated waters. These limits account for chronic risk, which describes their potential to induce health effects through the long-term exposure of consumers to a contaminant via drinking water. As discussed, known contaminants with regulatory limits in drinking water may be microplastics-associated. By knowing the adsorption capacities of contaminants to various types of plastic, it is possible to harness existing MCLs/MACs to develop preliminary guidance for managing microplastics in drinking water. As defined in the Safe Drinking Water Act, a contaminant is regulated in drinking water if (1) the contaminant may adversely affect human health, (2) it is highly likely that the contaminant will frequently be present in public water systems and at concentrations of public health concern, and (3) regulating the contaminant will result in the reduction of health risk for drinking water consumers.[53] When these criteria have been fulfilled, the health effects resulting from contaminant exposure through drinking water must be determined; this enables the calculation of an MCL/MAC.[54] Accordingly, not all chemical contaminants in drinking water are currently regulated, and MCLs/MACs are not available for some of them.

To facilitate advancement of the management of health risks attributable to waterborne microplastics in the water industry, even in the absence of comprehensive and conclusive toxicological data for the various microplastics themselves, the concept of the Threshold Microplastic Concentration (TMC) was developed. Recognizing that a quantitative health risk assessment of microplastics themselves is not yet possible due to the lack of requisite data, the TMC framework was used to identify (1) microplastic concentrations that may result in the intake of regulated contaminants on/in microplastics at levels of human health concern and (2) treatment targets for managing those potential risks. This potential health risk is addressed considering the worst-case scenario, in which the maximum amount of a single contaminant is sorbed to the microplastics and released in the human gut upon ingestion of water containing the microplastics. Worst-case scenarios are commonly used in risk assessment and management.[55] The developed framework is presented by using available data; however, it can be updated as more information becomes available.

2. METHODS

Regular identification and evaluation of all potential contaminants sorbed on microplastics in a given system are not practically feasible. Thus, a generally conservative approach was applied herein using currently available data to identify the TMC (i.e., waterborne microplastic concentration that may result in intake of regulated contaminants sorbed on microplastics at levels of human health concern) and treatment targets for managing those potential risks. In this section, (1) the approach to evaluating the TMC is described, (2) the different data including microplastics size, shape, and type, as well as the contaminants considered and their health guidelines that are used in the framework are discussed, and (3) the equations used to derive the TMC are presented.

2.1. TMC Concept and Evaluation Framework Development

To evaluate the TMC, six key data were collected, summarized, and integrated: (1) microplastic size, (2) microplastic shape, (3) microplastic polymer type, (4) health guidelines defined by MCLs or MACs, (5) contaminant adsorption capacity on plastics, and (6) the extent of microplastic removal during drinking water treatment (if

Table 1. Lowest Drinking Water Guideline and Highest Adsorption Capacity on Plastics for Contaminants Reflected in the TMC Framework

contaminant	adsorption capacity (mg/g)	adsorption capacity (mg/m^2)	microplastic type	microplastic size (μm)	adsorption reference	MCL or MAC (mg/L)[a]
aluminum	0.27[b]	185	PET	3000	67	2.9
antimony	27.8	5720	PC	1000	68	0.006
arsenic	1.12	20	PS	100	37	0.003
bromine	13	2330	PS	1000	35	0.04
cadmium	0.03	0.748	HDPE	154	69	0.005
chromium	0.000454[b]	0.31	PET	3000	67	0.03
copper	1.32	275	PVC	900	70	2
lead	0.00187[b]	1.28	PET	3000	67	0.005
manganese	0.13[b]	89	PET	3000	67	0.12
mercury	0.00125	0.0009	HDPE	4.5	73	0.002
polychlorinated biphenyls	0.35	247	PP	5000	74	0.0005

[a] MCLs and MACs were obtained from USEPA, Canadian, Australian and WHO guidelines.[62,63,51,52] [b] Amount desorbed after exposure to contaminant in the environment.

relevant). These were the minimum data requirements to create a framework and calculate the TMCs. Other factors (for instance, microplastic weathering/aging) can be integrated but were not considered in developing the TMC framework due to the lack of consistent, well-defined characteristics, as discussed in Section 3.4. Microplastics size and shape were required to reflect various surface areas for contaminant sorption. Microplastics types and adsorption capacities were necessary to evaluate the masses of contaminants that could be associated with these microplastics. MCLs or MACs were used because they are enforceable standards that are set to regulate contaminants in drinking water, which make them fit for inclusion in the TMC framework. The extent of microplastics removal during drinking water treatment, which varies across treatment units and plants, was included in the framework for TMC evaluation to link raw water microplastic concentrations and exposure through treated water. This parameter was included to facilitate site-specific analysis. Microplastics removal can be evaluated in treatment plants or estimated at pilot-scale. Information regarding microplastics removal during drinking water treatment was not synthesized herein to obtain values of this parameter.

Adsorption capacity is normally reported per unit mass of a particular size, shape, and type of microplastic, limiting its applicability to microplastics of different sizes and shapes. A core assumption of this work was that the adsorption capacity per unit of microplastic particle surface area should be the same even if the size and shape changes. Mass-based adsorption capacity values can be converted to surface-area-based values if the size, shape, and type of microplastics used in the adsorption capacity experiment are reported. Given a surface-area-based adsorption capacity and an MCL or MAC of contaminant, it is possible to back-calculate the concentration of any microplastics size and shape with contaminant adsorbed to full capacity that results in contaminant concentrations equal to the MCL or MAC—the TMC. This framework can then be used to evaluate if a given microplastics concentration, size, and shape of concern in a particular water supply, combined with the adsorption capacity for contaminants of concern, constitutes a potential risk. Currently, microplastics in environmental samples are commonly identified visually using microscopy and analytical techniques such as spectroscopy, and they are typically reported as count-based concentrations,[56] so the TMC is also expressed as a count-based concentration. If required, the physical properties of microplastics such as size, shape, and density can be used to estimate mass concentrations.

2.2. Inputs Used for Calculating TMCs

In this study, a TMC was calculated for each of several sizes and shapes of microplastics as well as for each of several contaminants.

Microplastics ranging from less than 1 to 5000 μm have been found in freshwaters used as sources for producing drinking water.[57] Here, a minimum particle size of 1 μm was used to stay within the size range defined for microplastics; particles smaller than 1 μm fall in the size category of nanoplastics.[58] Even though particles larger than 100 μm are generally well removed by physicochemical filtration,[59] an upper particle size cutoff of 750 μm was arbitrarily selected to analyze the effect of different sizes. Particle size was defined herein as the longest dimension of the particle.

Waterborne microplastic shapes depend on their source. Primary microplastics have a defined shape because they are manufactured according to specifications for their applications (e.g., cosmetic "microbeads"). Microplastics shapes also vary due to weathering[60] and result in the formation of smaller secondary microplastics of irregular shapes. A synthesis of over 100 investigations in which microplastics were characterized indicated that fibers and fragments are among the most commonly detected microplastics in the environment.[61,62] Accordingly, shapes representing fibers and fragments were analyzed herein, including cylinders (short, long, and with equal diameter and height), oblate spheroids (ellipticity of 0.2 and 0.9), spheres, and cubes (Table S1). Of course, additional shapes and associated surface areas could be introduced for sensitivity analysis or additional conservatism if warranted.

Initially, high density polyethylene (HDPE), low density polyethylene (LDPE), polyethylene terephthalate (PET), polypropylene (PP), polystyrene (PS), and polyvinyl chloride (PVC) were considered in the TMC framework development because they are the most common waterborne microplastics.[60] Polycarbonate (PC) was also considered in this analysis because it has commonly been found in wastewater along with PET.[62,64] Because adsorption capacity data were not available for each contaminant−plastic combination, the effects of microplastics/polymer type were ultimately excluded from this analysis. The highest available adsorption capacity value for each contaminant was used and applied to all microplastics/polymer types.

Drinking water guidelines and/or standards were used to identify health-based thresholds for the chemical contaminants adsorbed to the microplastics—below these levels, there is no known or expected risk to health.[51,52,65,66] Daily water intake and contaminant exposure levels are reflected in the MCLs and MACs for drinking water contaminants.

Literature-acquired adsorption capacities were used to indicate the greatest extent of potential chemical contaminant delivery from microplastics. These values were obtained from adsorption isotherm studies (Table S2). For cases where multiple adsorption capacities were reported for a contaminant, the highest among them per unit of surface area was selected to reflect the worst-case scenario (Table 1). Due to limited adsorption capacity data, one experiment[67] was included that provides the amount of contaminant desorbed after exposure in the environment rather than the maximum amount of contaminant that can be adsorbed; the full adsorption capacity could

Table 2. Literature-Acquired Data and Assigned/Computed Model Inputs Used to Calculate the TMC[a]

inputs	
literature-acquired	assigned/computed
microplastics size used in adsorption experiments (μm)	size range considered in TMC calculation (1−750 μm)
adsorption capacity ($AC^{*}_{\text{mass},\,px}$, mg/g)	shape (short cylinders, long cylinders, and cylinders with equal diameter and height, oblate spheroids with ellipticity values of 0.2 and 0.9, spheres, and cubes)
density of microplastics type used in adsorption experiment (ρ_p, g/m^3)	surface area of one particle in adsorption experiment and considered in TMC calculation: $A^*$and A_{ij} (m^2), respectively
drinking water guideline/standard value (H_x, mg/L)	volume of one particle in adsorption experiment, V^* (m^3)
	specific surface area per gram of microplastics in adsorption experiment, SSA^{*}_{p} (m^2/g)
	estimated adsorption capacity per unit surface area, $AC_{\text{SA,px}}$ (mg/m^2)

[a] i = size of microplastics; j = shape of microplastics; p = type of microplastics (HDPE, LDPE, PC, PET, PP, PS, PVC); x = contaminant.

thus be higher than stated. It should be noted that Table 1 is not an exhaustive list of all contaminants that may be associated with microplastics. It lists only those contaminants for which all the data required to evaluate the TMC were available at the time of conducting this assessment and, hence, is not fully representative of contaminants of all types of adsorption characteristics. This is because the adsorption capacity of many regulated contaminants to different microplastics has not been reported, thereby precluding a comprehensive assessment here. Nonetheless, the availability of some data enabled the development of a framework that can be updated as more data become available so that more contaminants or specific microplastic types can be reflected in the TMC calculations. The data used here are for the purpose of demonstrating (1) how the framework can be used to evaluate potential health risks and (2) interpretation of results obtained from the analysis.

2.3. Calculation of the TMC

Adsorption capacities of contaminants on microplastics are reported on a mass basis in the literature (i.e., mass of contaminant per unit mass of microplastics, typically in mg/g). Surface-area-based adsorption capacities were derived by using these values and the size, shape, and type of microplastics used in the adsorption experiments. This conversion involves computation of the amount of surface area per gram of microplastics to which the reported adsorption capacity corresponds. Adsorption experiments typically use commercially available "microspheres", which are essentially spherical microplastics. Additionally, spheres have the smallest amount of surface area per unit mass and thus may give a conservatively high estimate of the adsorption capacity per unit surface area. Assuming conditions to reflect a comparatively high adsorption of contaminants enable the TMC calculation to be protective. The surface area of one microplastic particle of the size and shape of microplastics considered in the TMC analysis is then calculated (Table S2). Monolayer coverage was assumed to extrapolate the adsorption of contaminants to other sizes and shapes of microplastics because experimental data of contaminant adsorption capacities on different shapes and a range of sizes are currently unavailable. Finally, the adsorption capacity per unit surface area and the surface area per particle are used to compute the minimum number of such microplastics per liter of water required for the adsorbed contaminant concentration to reach the MCL/MAC.

Mathematically, the TMC was calculated according to eq 1, in which TMC_{ijpx} is the concentration of microplastics of size i, shape j, and type p that can carry an amount of contaminant x equal to the MCL/MAC (H_x). $AC_{\text{SA},px}$ is the converted adsorption capacity per unit surface area for a particular microplastic type and contaminant, and A_{ij} is the surface area of the microplastic considered. The extent of treatment is represented by log-removal T so that $T = 1$ corresponds to 90% removal of particles, $T = 2$ corresponds to 99% removal, and so on. The treatment exponent is positive because the higher the removal of microplastics particles is, the higher the TMC can be in the source water without exceeding the health guideline.

$$\text{TMC}_{ijpx} = \frac{H_x}{\text{AC}_{\text{SA},px} \times A_{ij}} \times 10^{T} \tag{1}$$

$AC_{\text{SA},px}$ is calculated using eq 2, in which $AC^{*}_{\text{mass},px}$ is the literature-based adsorption capacity per unit mass of microplastics type p and contaminant x based on the adsorption capacity experiment in the literature (*). In the absence of type-specific data, the $AC^{*}_{\text{mass},px}$ of a given contaminant was assumed to be the same for all microplastics types. SSA^{*}_{p} is the specific surface area defined as the total surface area per gram of the size, shape, and type of microplastics used in the adsorption experiments. It is calculated using eq 3, in which A^* and V^* correspond to the area and volume of a single microplastic particle based on the size and shape used in the adsorption experiment, respectively, and ρ_p corresponds to the density of microplastic type p.

$$\text{AC}_{\text{SA},px} = \frac{\text{AC}^{*}_{\text{mass},px}}{\text{SSA}^{*}_{p}} \tag{2}$$

$$\text{SSA}^{*}_{p} = \frac{A^{*}}{V^{*} \times \rho_p} \tag{3}$$

The above steps were performed for all of the combinations of particle size and shape as well as contaminant to evaluate the sensitivity of the TMC to changes in these input values. The inputs used in the TMC calculations are summarized in Table 2. TMC was calculated for each of 11 contaminants, seven shapes, and a range of longest dimensions. A sample series of calculations to determine a TMC is provided in the Supporting Information.

3. RESULTS AND DISCUSSION

3.1. Evaluating the TMC

The TMC, indicating the total number of microplastics per liter of treated or untreated water that may result in exposure to potentially harmful concentrations of chemical contaminants on/in microplastics if ingested, was computed for a wide range of microplastic sizes and shapes as well as contaminants. Before the results are presented, it is critical to correctly interpret what the TMC reveals. As the TMC is threshold-based, it may seem analogous to MCLs or MACs that are used to manage drinking water risks for chemical contaminants in the United States.[51] and Canada,[52] respectively. However, unlike MCLs and MACs that indicate the maximum acceptable concentration of a contaminant in drinking water, the TMC only indicates a microplastics concentration that *may* indicate potential health risk if exceeded. This is because the amount of contaminants actually present may be less than the adsorption capacity (i.e., higher amounts of microplastics may not pose a significant risk if only small amounts of contaminants are adsorbed). Therefore, if the microplastic concentration in a

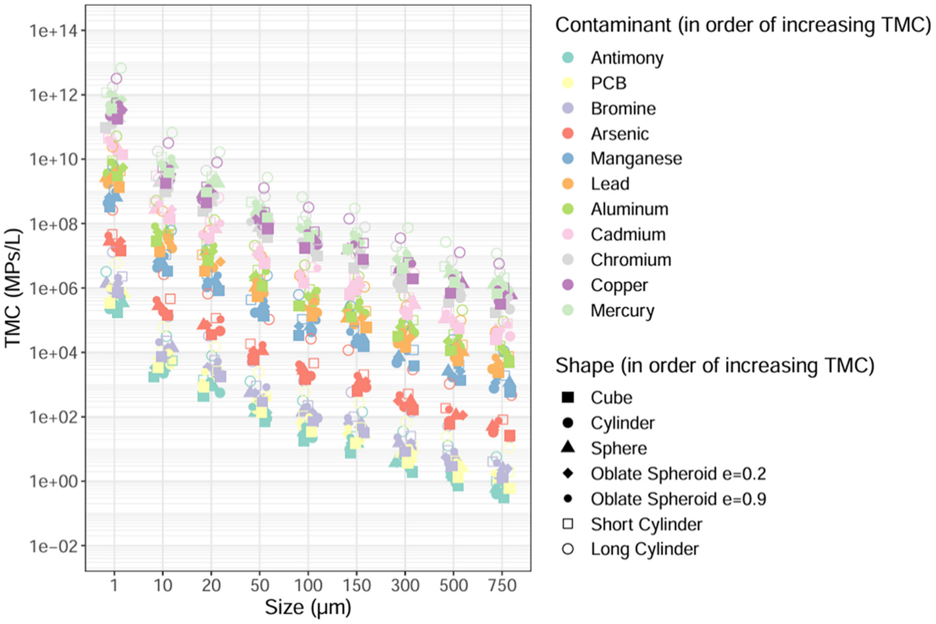

Figure 1. TMC is presented for all combinations of microplastics size and shape as well as contaminants currently included in the framework. For each particle size, TMC values are horizontally jittered to facilitate visualization.

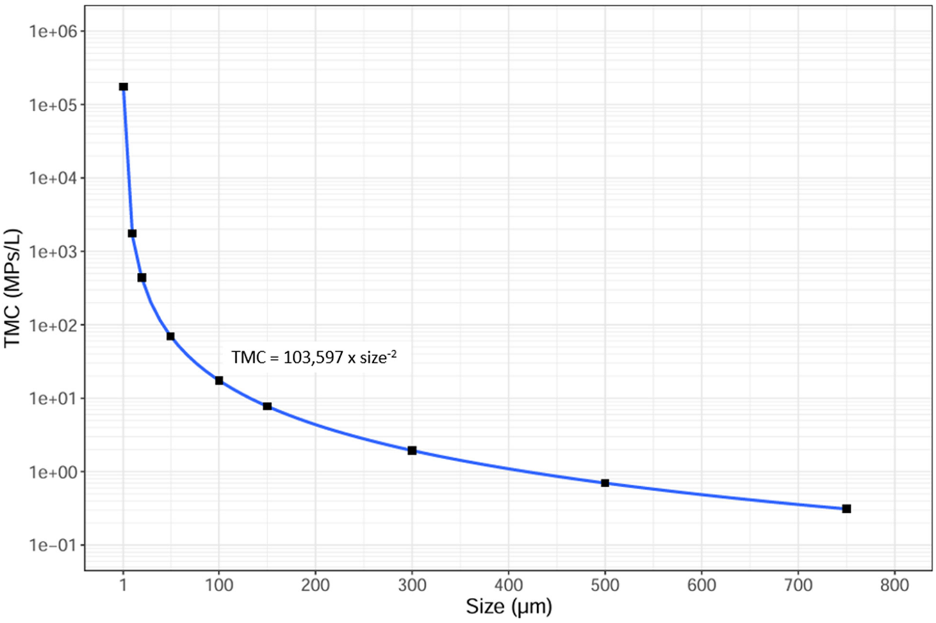

Figure 2. Curve with the lowest TMC values in the current framework (generated for antimony sorbed on cube-shaped microplastics) is shown.

water sample is lower than the TMC (calculated based on a worst-case scenario approach), the water can reasonably be considered safe or not requiring additional treatment for managing risk associated with adsorbed contaminants. If the microplastics concentration exceeds the TMC, this does *not* necessarily indicate an immediate health risk and the need for additional treatment; rather, it highlights that a potentially concerning concentration of microplastics is present and warrants further assessment of water quality and health risk. When TMC values are compared for different scenarios, it is important to recall that a lower TMC corresponds to higher potential for risk because fewer microplastic particles are needed for contaminant exposure at concentrations associated with health impacts.

3.1.1. Sensitivity of the TMC to Changes in Microplastics Size and Shape. In Figure 1, TMCs calculated for all combinations of contaminants and microplastic size and shape are presented to illustrate how these different factors affect TMC, which ranges over about 12 orders of magnitude. Different sizes are represented on the *x*-axis, the spread of colors within a size represents the effect of different contaminants on the TMC, and the spread of point types of a specific color within a size represents the effect of shape on the TMC. As seen from the distribution of plotted points, the major drivers of the TMC are microplastics size and contaminant type.

TMCs decreased as particle size and the resulting surface area per particle increased (Figure 1). When other factors (i.e.,

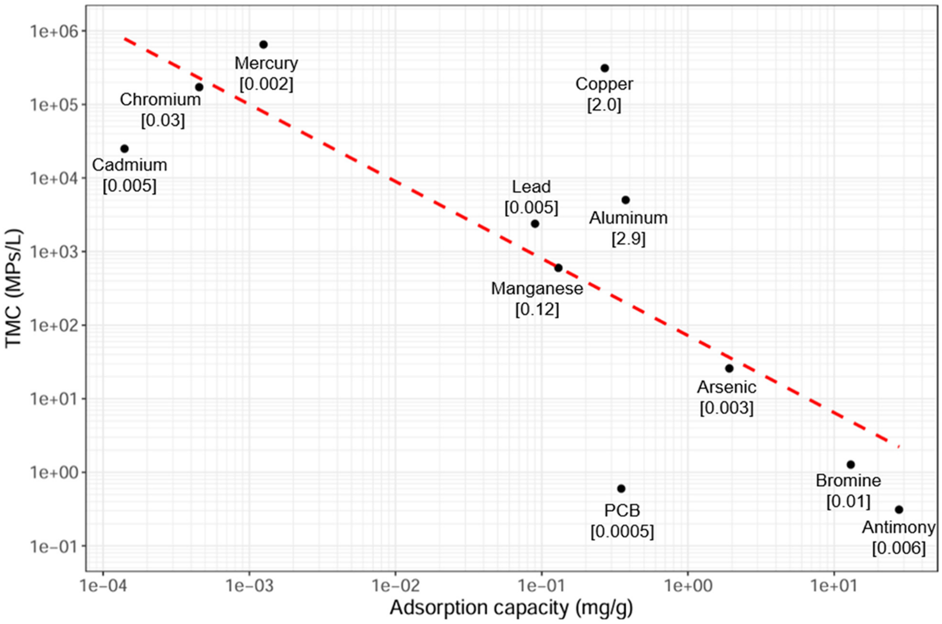

Figure 3. Relationship between TMC and adsorption capacity. TMCs for cube-shaped 750 μm microplastics are presented here. Values in square brackets are contaminant MCLs/MACs (mg/L).

shape, contaminant) were kept constant across the range of sizes analyzed, the TMC had a range of about 6 orders of magnitude, highlighting microplastics size as a major driver of TMC. The highest and lowest TMCs were thus obtained with 1 and 750 μm microplastics, respectively. Figure 1 shows that the lowest, potentially most concerning, TMCs were calculated for cube-shaped microplastics having a size of 750 μm that are contaminated with antimony. Other combinations of factors can be analyzed, but the most concerning combination of factors was chosen for the purpose of discussion. Figure 2 shows the curve of TMC values as a function of size by using the lowest (most concerning) TMCs from among all combinations of factors across the size range analyzed in this study (1−750 μm). This indicates how more particles are needed to adsorb a given amount of contaminant as the size of the microplastics gets smaller.

In contrast, microplastics shape only affected TMC over about 1.25 orders of magnitude of range (Figure 1). Notably, the lowest and highest TMCs in the presented analysis were calculated for cubes and long cylindrical particles, respectively. For all cases, the TMCs from lowest to highest based on shape followed the order cube < cylinder (diameter = height) < sphere < oblate spheroid (e = 0.2) < oblate spheroid (e = 0.9) < short cylinder < long cylinder.

With an increase in surface area per particle due to increasing size or difference in shape, the TMC decreases because more area is available for contaminant adsorption, and hence, a higher mass of contaminants can be associated per particle. This means that fewer particles with the contaminant adsorbed at full capacity can carry the contaminant at concentrations exceeding the MCL/MAC compared with particles with a lower surface area per particle. For all cases, the lowest TMCs were calculated for cubic microplastics of 750 μm size because this combination results in the highest surface area per particle and, hence, the capacity to adsorb more contaminants with fewer particles. Cubes were included in the TMC framework because, relative to the other shapes and for a given particle size (i.e., longest dimension), they have a larger surface area available for contaminant adsorption per particle relative to the other shapes included in the framework. Thus, they serve as a worst-case scenario that yields relatively more conservative (i.e., lower) TMC values. Realistically, it is unlikely to encounter cube-shaped microplastics in the environment.

Chemical effects of microplastics type on the TMC are not reflected here due to the incomplete data addressing adsorption capacity variation among plastic types. To develop the TMC framework with the amount of adsorption capacity information currently available, it was necessary to assume that the adsorption capacities were the same among microplastics types. Theoretical comparisons of adsorption capacities of contaminants on different types of microplastics can be made from an understanding of their chemical properties, such as charge density, polarity, and hydrophobicity.[71] Nonetheless, additional experimental data are necessary to provide a basis for a detailed quantitative discussion regarding the implications of different microplastics types.

3.1.2. Contaminant. The effect of different contaminants on the TMC is driven by both the toxicity and adsorption capacity on microplastics. For all cases, a combination of higher adsorption capacity and higher toxicity (i.e., lower health guideline concentration, such as MCL or MAC) of a contaminant resulted in a lower TMC when all other factors were constant (Figure 3).

For a given shape and size, the TMC has a range of about 6 orders of magnitude (Figure 1), highlighting contaminant type as a major driver of the TMC. Therefore, based on the TMCs calculated, the types of contaminants sorbed on microplastics were found to be a key driver in evaluating potential health concerns from microplastics. At present, sufficient data are not available to develop a reliable model relating TMCs to adsorption capacities of contaminants, and the trend shown in Figure 3 is relevant only for discussing results specific to the analysis presented in this study. Nonetheless, as more data become available, reliable models may be developed to calculate TMCs using the adsorption capacity of contaminants.

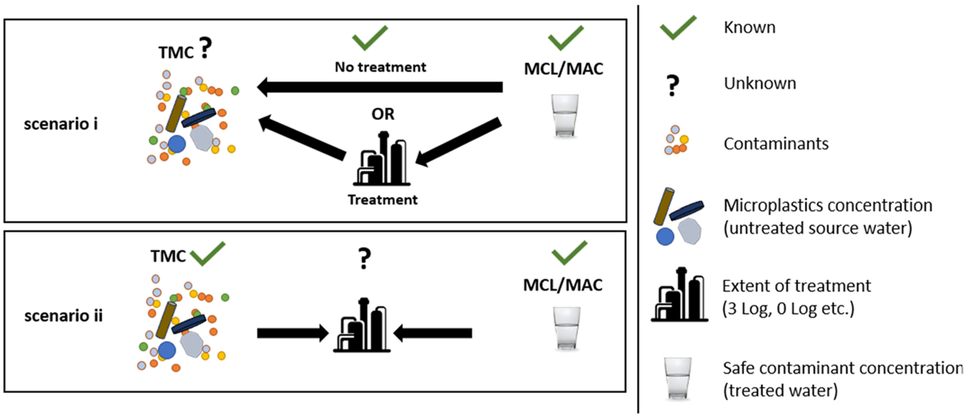

Figure 4. Different scenarios in which the TMC calculation framework can be used to evaluate the unknowns in the drinking water pathway.

3.2. Application Potential of the TMC Framework and Interpretation of Results

Water providers can use the TMC framework to inform the risk management needs associated with microplastics in source water. Ideally, this would be done based on source water microplastics characterization. First, microplastics should be extracted from the source water. They should then be enumerated, and the representative sizes, shapes, and polymer types should be characterized. These inputs can be entered into the TMC framework (with all possible contaminants for which adsorption data are available included). If the measured microplastics concentration exceeds the calculated TMC, further analysis is warranted to identify which contaminants are relevant for inclusion in the calculation of a site-specific TMC. For example, based on available data, antimony drives TMC in the examples above; however, if it is not present in source water, it can be excluded from TMC calculation. Hence, the presence of antimony and sufficiently high concentrations of microplastics in source water indicate the need for further analysis to confirm the adequacy of treatment (i.e., antimony removal in this example).

At present, ideal source water microplastic characterization is rarely possible. In the absence of those data, the TMC framework allows a conservative, worst-case scenario approach to assessing potential threats from waterborne microplastics by using maximum adsorption capacities (i.e., presuming spheres while deriving surface area-based adsorption capacities and including all contaminants for which adsorption data to plastics are available). As contaminant adsorption data or site-specific microplastics source water characterization data become available, TMC analysis can be refined. Nonetheless, taking a worst-case scenario approach is an essential starting point for preparing for potential challenges and mitigating risks. Although microplastics in drinking water are not yet regulated, the consideration of worst-case scenarios underscores the need to develop standard methods for their identification and enumeration. While some methods have been reported, they are not reliable, precluding regular monitoring.[72] While strategies for managing microplastics in drinking water have emerged and include methods guidance,[76] uncertainty in concentration estimates has yet to be addressed, though statistical approaches for evaluating uncertainty in enumeration-based concentration estimates and interpretation of nondetects of discrete particles are available.[77,78]

As data become available, the application of the TMC framework can be increasingly site-specific. Primarily, the results of the analysis can provide an estimate of whether any given concentration of microplastics in source water constitutes potential risks due to the adsorbed contaminants. However, it can also be used to calculate either the source or treated water microplastics concentration above which consumption of drinking water may result in the ingestion of regulated contaminants sorbed on microplastics at levels of human health concern or the extent of treatment required to produce potable water from a source water with a known microplastics concentration (Figure 4).

Scenario (i) involves the back-calculation of a permissible source water microplastic concentration (i.e., TMC) given known levels of treatment and health guidelines for relevant contaminants so that a measured source concentration can be compared against it. To evaluate the minimum extent of treatment required in scenario (ii), the source microplastics concentration is set equal to the TMC and the required treatment is back-calculated using the MCLs/MACs and eq 1. Whether considering the TMC in the treated water or untreated source or determining the extent of treatment required, it is desirable to have a measured waterborne microplastics concentration that is below the lowest TMC value that is evaluated on a case-by-case basis (i.e., by considering microplastics size, shape, polymer type, and contaminants relevant to the specific system).

3.3. Potential for Advancement of the TMC Framework to Microplastics Hazards Other than Adsorbed Contaminants

The TMC framework could eventually be implemented to account for the different ways in which microplastics are potentially a health concern in drinking water; the TMC as a result of the microplastic particles only ($\text{TMC}_{\text{MP-P}}$), the TMC due to the microplastics additives ($\text{TMC}_{\text{MP-A}}$), and the TMC due to adsorbed contaminants on microplastics ($\text{TMC}_{\text{MP-C}}$). Each of these components can drive the TMC depending on which of them is lowest (i.e., highest potential for risk). Therefore, the overall TMC would be the lowest of these components: $\text{TMC}_{\text{Overall}} = \min(\text{TMC}_{\text{MP-P}}, \text{TMC}_{\text{MP-A}}, \text{TMC}_{\text{MP-C}})$. Currently, there are not sufficient data to quantify risks to human health posed by the microplastics themselves and derive a drinking water MCL or MAC. Plastic additives are known to have toxic properties, but exposure to them and an acceptable level of risk through drinking water ingestion need to be determined before their MCL/MAC can be calculated. Several uncertainties complicate calculation of $\text{TMC}_{\text{MP-A}}$ at present; these include additive leaching rates and their driving factors in both source water and the human body. Hypothetically, if 1 μg of additive is present in 1 g of microplastic, will all of the additive mass leach to the human gut or water in the distribution system? How quickly? Of course, immediate and complete release of the additive can be assumed as a worst-case scenario. Given the lack of data to derive $\text{TMC}_{\text{MP-P}}$ and $\text{TMC}_{\text{MP-A}}$, a starting point for assessing $\text{TMC}_{\text{Overall}}$ can be based on the effect of sorbed contaminants, for which sufficient data are available to develop a framework and conduct exploratory assessments for preparing for potential challenges and mitigating risks.

3.4. Conceptual Model Limitations

Ideally, calculation of a TMC for adsorbed contaminants would require (1) the sizes, shapes, and types of waterborne microplastics to be exactly known, (2) the concentration of all possible contaminants adsorbed on the microplastics to be known, (3) the adsorption capacities of those contaminants to the different microplastic types and the desorption characteristics from microplastics in the human gut to be understood and describable, and (4) an MCL/MAC for each contaminant to be available. In such a case, the TMC would be easily evaluated if waterborne microplastic concentrations and associated contaminant concentrations could be accurately measured. In practice, however, this is not possible because monitoring of waterborne microplastics is challenging,[17,75] adsorbed contaminants are varied, and regular full-suite contaminant monitoring is cost-prohibitive. Knowing which contaminants are present in the source water and their adsorption capacities on microplastics is integral to evaluation of the TMC. Several simplifying assumptions were required to enable TMC evaluation, as discussed below. The assumptions made in this study are, however, not characteristic of the framework itself; they were necessary to conduct the assessment only in the absence of required data. As more information becomes available, fewer assumptions will be required.

In this TMC framework, the entire microplastic surface area was assumed smooth and available for contaminant sorption. As mentioned earlier, monolayer coverage was also necessarily assumed. This was required to estimate adsorption capacities of contaminants on microplastics of all shapes and sizes analyzed. While microplastics that are porous or have extremely rough surfaces would have greater surface area, it is impossible to characterize these surfaces comprehensively or *a priori*. It is also unlikely, however, that microplastic surfaces are entirely covered by an individual contaminant, and the surface area available for one contaminant may be reduced by the presence of others. Assuming that the contaminant is sorbed at full capacity on microplastics and that the entire contaminant mass desorbs from the microplastics and is bioavailable upon ingestion likely leads to the calculation of lower, more concerning TMCs. If warranted, additional particle shapes can be considered to adjust surface areas (and hence TMCs) or scaling factors can be introduced to adjust adsorption−desorption capacities to reflect surface areas of environmentally relevant microplastics. For example, scaling factors could be added to eq 1 to reflect that complete contaminant desorption is not expected in invertebrate (and potentially human) gut environments.[74] However, the development of these scenarios and scaling factors is beyond the scope of the present investigation.

In cases where adsorption capacity data for a given contaminant were available for only one type of microplastic, adsorption was necessarily assumed to be the same for other types of microplastics. Even though adsorption is influenced by polymer type and contaminant chemical properties, this assumption is necessary to enable development of the framework in the absence of experimental data on adsorption capacities of all combinations of contaminant and microplastics type. The use of adsorption capacity values derived from laboratory experiments may also lead to a potential overestimation of exposure to chemical contaminants on microplastics because microplastics in these experiments were exposed to high contaminant concentrations. This does not necessarily reflect environmental conditions, which may include competition for surface sites and contaminant desorption into the water column. Additionally, the TMC calculation assumes that the MCLs/MACs of contaminants are reached solely through microplastics, but contaminants may also be present in the water matrix and may potentially be associated with other particles.

The TMC calculations in this study presume that the considered microplastics are of uniform size and shape; however, environmental microplastics are expected to be more diverse.[1,2] Analysis of the effect of diverse microplastics sizes, shapes, and types with differing adsorption capacities was beyond the scope of this study. Notably, MCLs and MACs also do not consider synergistic effects resulting from the concurrent presence of multiple hazards.

In addition to the six key data explored in this study that were necessary to develop a flexible framework to calculate TMCs, additional factors can be included as required to expand the analysis. Factors such as weathering or aging can influence physical or chemical properties of microplastics (e.g., surface cracks or addition of functional groups) which may affect their contaminant adsorption capacities.[79,80] They could potentially be integrated into the TMC framework by introducing scaling factors in eqs 2 and 3; however, the effects of weathering (on microplastics size, shape, total surface area, surface charge, internal surface area, contaminant adsorption/desorption capacity, etc.) would need to be characterized first. Representation of environmental conditions and weathering times is particularly challenging because they are site-specific and highly variable.

Despite the lack of ideal knowledge regarding microplastics and associated chemicals in water supplies used for potable water production, a science-based approach for risk management can be developed for use by water providers. The described TMC concept has been used to develop a simple, updatable framework to calculate TMCs for various operational scenarios so that the lowest (and likely most conservative) of the calculated TMCs can be used for decision-making. It must be re-emphasized that the purpose of the TMC framework is to indicate whether existing treatment is sufficient for managing potential health risks attributable to ingestion of contaminants sorbed to microplastics in drinking water based on currently available science or if more detailed risk assessment is needed—the framework is *not* designed to quantitatively evaluate health risk. Given the necessary assumptions discussed above and that contaminants may not be present at full capacity on microplastics, use of the framework to calculate a TMC to decide if enhanced drinking water treatment is required is strongly discouraged in the absence of considerable additional analyses (e.g., site-specific microplastics characterization).

While the present framework focuses on evaluating TMCs for combinations of a single particle shape and contaminant type, it can be expanded to reflect new insights regarding key assumptions, water contaminants of health relevance, additives released from microplastics, health guidelines (e.g., MCL/MAC), heterogeneous mixtures of contaminants in water, and health impacts of microplastics themselves as more toxicological data become available. At present, the investigation has focused on using available data to represent a worst-case scenario for evaluating adsorbed contaminant TMCs that can be used to inform management strategies to deal with microplastics in water. Unlike MCLs and MACs that indicate the maximum acceptable concentration of a contaminant in drinking water, the TMC only indicates a microplastics concentration that *may* indicate potential health risk if exceeded. Nonetheless, a worst-case scenario approach such as this can be useful for water providers in identifying when a certain hazard, like contaminants adsorbed to microplastics that were discussed here, is not likely to pose a significant health threat requiring management intervention. Thus, the TMC framework presented here can help to advance the water industry's philosophy and approaches for quantifying microplastics in source water, managing threats from microplastics, and monitoring microplastics more broadly.

4. CONCLUSIONS

Several conclusions based on the evaluation of the potential health risks from contaminants sorbed on microplastics in drinking water were drawn from this work. They include:

- A new concept called the threshold microplastics concentration (TMC) is useful to indicate the total number of microplastic particles per liter of untreated source water or treated water that *may* constitute exposure to potentially harmful concentrations of chemical contaminants retained on microplastics if ingested. Lower TMCs correspond to higher potential for risk because they indicate that fewer microplastic particles might lead to contaminant exposure at concentrations associated with health impacts. If the microplastics concentration for a given system is lower than the TMC, it can be considered safe with respect to adsorbed contaminants based on all the conservative assumptions in the calculation of the TMC and the set of contaminants considered. However, exceeding the TMC does not necessarily indicate greater health risk from the considered contaminants than drinking water at the MCL/MAC; rather, it warrants further investigation of water quality and treatment processes.
- The TMC concept and framework consider a worst-case scenario approach when using the inputs/assumptions made in calculations. It provides utilities and regulators with a science-based, easily updatable approach for screening drinking water systems to indicate if existing treatment is sufficient for managing potential health risks attributable to contaminants adsorbed to waterborne microplastics or if more detailed risk assessment is needed.
- The TMC framework uses available microplastics and contaminant data, and it has been modularized such that it can be updated as more information regarding toxicity (i.e., MCLs/MACs) and sorption capacities of various combinations of contaminants and microplastics becomes available. It can also incorporate expected treatment efficiency. Thus, it can be applied for system-specific risk management to calculate (1) the source water microplastics concentration above which consumption of downstream treated water may result in the ingestion of regulated contaminants sorbed on microplastics at levels of human health concern or (2) the extent of treatment required to produce potable water that does not result in the potential ingestion of regulated contaminants sorbed on microplastics at levels of human health concern.
- For all cases analyzed based on microplastics shape, the sequence of lowest to highest TMCs was: cube < cylinder (diameter = height) < sphere < oblate spheroid (e = 0.2) < oblate spheroid (e = 0.9) < short cylinder < long cylinder. In the present analysis, TMCs calculated for cube-shaped microplastics with antimony on their surfaces resulted in the lowest, most-conservative TMCs of any size. For example, these TMC values calculated for 10 and 100 μm microplastics are 1748 and 17.5 microplastics/L, respectively. If the source water microplastics concentration is higher than the TMC calculated using a reference size, further water quality analysis is recommended. The TMC, however, should not be considered as an enforceable standard without confirming that contaminants are adsorbed at their full capacities.
- Based on currently available data, the present analysis demonstrates that meaningful targets for monitoring source water microplastics concentrations can be established using the TMC framework, and it can be updated as more information regarding adsorption of contaminants on microplastics and their toxicology becomes available.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/envhealth.3c00174.

Example TMC calculation, details of microplastics shapes and geometry, adsorption capacities of contaminants on microplastics, and physical characteristics of microplastics in aquatic environments (PDF)

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Notes

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