

Majority of potable water microplastics are smaller than the 20 μ m EU methodology limit for consumable water quality

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RESEARCH ARTICLE

Majority of potable water microplastics are smaller than the 20 µm EU methodology limit for consumable water quality

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Abstract

Microplastic (MP) content in nutrition including potable water is unregulated, although MP concentrations in individual brands of bottled water diverge by several orders of magnitude. The EU Directive 2020/2184 on consumable water quality recently proposed methodological approaches to the detection of MPs in potable water in the size range of 20-5000 µm. However, fine MPs in the 1–20 µm range are far more likely to pass the human intestine into blood and organs. To evaluate the impact of this omission on the total number of detected MPs, we determined MP concentrations down to 1 µm in ten different brands of polyethylene terephthalate (PET) bottled water and one tap water sample using automated Raman microspectroscopy. We found that MP concentrations ranged from 19 to 1.154 (n/L) [0.001 to 0.250 µg/L], and although all the investigated samples of bottled water were stored in PET containers, PET accounted only for a small percentage of identified MPs in most samples. Importantly, 98 and 94% of MPs measured less than 20 and 10 µm in diameter, respectively, demonstrating the importance of small MP inclusion in potable water analyses and regulation. The current study presents a protocol to identify MPs down to 1 µm in any type of potable water regardless of hardness, and demonstrates the importance of implementing both negative and positive procedural, guality control measures.

1. Introduction

A 'Microplastic' (MP) particle is defined as any plastic item which longest axis measures between 1 to 5000 μ m (5 mm) [1, 2], or 1 to 1000 μ m (1 mm) according to the International Organization for Standardization (ISO) [3]. MPs are ubiquitous pollutants [4] and have inevitably made their way into our nutrition [5] and potable water supply; both tap and bottled [6]. Bottled water especially, has been scrutinized for MP contamination [7, 8] and on a global, annual base, we consume more than 60 L of bottled water per capita [9].

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Although it has been demonstrated, and reconfirmed in the current study, that MP concentrations in individual brands of bottled water diverge by several orders of magnitude [7], no certification, regulations or market guidelines for MP contamination in potable water currently exist. However, the European Commission recently published a directive (2020/2184) on methodology to measure MPs in water intended for human consumption [10]. The directive accepts a limit of detection of 20 µm despite MPs <10 µm being considered to pose the greatest risk to human health [11]. To date, most scientific studies on MP content in drinking water have omitted the 1–20 µm size range due to detection limitations [12]. Yet, the limited number of studies that have investigated the presence of MPs down to 1 µm in potable water, suggest that MPs <20 µm are significantly more abundant [13, 14]. The widespread omission of the 1–20 µm size range in potable water research may have influenced the European Commission's decision to set the detection threshold at 20 µm.

Given this limitation, it is crucial to establish a standardized approach to measure and classify MPs down to 1 μ m in potable water before enacting potential regulations. To this end, we established a comprehensive protocol to determine MP content in any type of potable water regardless of hardness. To demonstrate the applicability of the protocol, ten different brands of bottled water and one tap water sample were investigated. We employed Raman microspectroscopy in combination with dedicated computer vision post-processing (Microplastic Solution, France) to correct for unintentional, visual particle partitioning and to more efficiently sort the data. In total, more than half a million individual particles were investigated of which n = 1,824 were identified as synthetic polymers. We found concentrations ranging from 19 to 1,154 MPs (n/L) [0.001 to 0.250 µg/L] compared with municipal tap water at 413 MPs (n/L) [0.096 µg/L] following procedural blank- and recovery corrections. In addition to demonstrating the application of the current protocol, this study highlights the importance of small MP inclusion in potable water analyses and regulation.

2. Methods and materials

2.1. Laboratory pre-treatment

In ten different French, anonymous brands of bottled water, MP ($\geq 1 \mu m$) concentrations were established by automated Raman microspectroscopy. Additionally, one potable tap water sample, representing the Toulouse Metropole area, was collected directly from the tap into five individual 1L glass bottles in our dedicated MP laboratory, and later examined. For all samples, 4.5 L of water was filtered through hydrophilic 0.45 μm , 47 mm polyvinyl fluoride (PVDF) filter membranes (Durapore, Merck KGaA, Germany), using a glass vacuum filtration device.

Due to buildup of residue on the filter membrane, hindering automated Raman microspectroscopic analysis, the filtered content underwent acidic and oxidative digestion treatment prior to analysis. Each filter membrane was transferred into 50 mL glass vials with 30 mL 30 vol.% hydrogen peroxide (H₂O₂) (Fischer Scientific, Belgium). To improve the transfer from the filter membrane to the H₂O₂-solution, each vial and its contents were ultrasonicated (BPAC, France) for 1 minute. Consequently, the filter was evacuated from the vial using a stainless-steel tweezer while being flushed with H₂O₂ (30 vol.%) to impede particles from sticking onto the filter membrane, thus improving MP recovery. The residual content of the filter was flushed with H₂O₂ (30 vol.%) into the vial until it held 40 mL. Following six continuous days of hotplate-induced digestion at 50°C in 40 mL H₂O₂ (30 vol.%) solution [15–17], 5 mL of 5 vol.% hydrochloric acid (HCl) was added to each vial to increase the acidity of the solution to incite the disintegration of carbonate residuals [18]. The reagents together lead to a redox reaction that is weaker than Fenton's reaction [19], slowly forming hypochlorous acid (HOCl) and water (H₂O) [20]. Following 24 h of reaction, all samples were filtered through individual $0.2 \,\mu$ m, 25 mm aluminium oxide filter membranes (Whatman Anodisc, U.K.) and flushed with ultrapure grade-A milliQ water (18.2 M Ω ·cm), leaving the desired particles on a flat surface suitable for microspectroscopic Raman analysis [21].

Prior to use, all chemical reagents were filtered through 0.45 µm PVDF filter membranes and all glassware was kiln sterilized at 500 °C for one hour to subsequently be fully covered in aluminium foil. All sample manipulation was carried out inside a FlowFAST V12P laminar flow cabinet located within a dedicated MP laboratory in Toulouse, France. In addition, red cotton lab coats were worn at all times inside the laboratory. No sampling permits were required as the bottled water was purchased from a local supermarket and the municipal tap water was sampled directly from the laboratory tap.

2.2. Raman microspectroscopy

For each sample, three individual 2x2 mm grid subsamples were examined, corresponding to 7.8% of the total filtered area. In total, n = 660,683 particles were investigated, averaging n = 55,057 particles per sample (~18 thousand particles per 2x2 mm grid subsample), including the procedural blank. All particles measuring $\geq 1 \, \mu m$ in area-equivalent diameter (circular model) were subject to Raman analysis. Raman measurements were carried out at 20°C using a Horiba LabRAM Soleil (Jobin Yvon, France). The samples were excited at 8% (7.2 mW) power output with a high stability air-cooled He-Cd 532 nm laser diode utilizing a Nikon LV-NUd5 100x objective. The lateral resolution of the unpolarized confocal laser beam was on the order of 1 µm. Spectra were generated in the range of 200–3400 cm⁻¹ using a 600 grooves/ cm grating with a 100 μ m split. The spectral resolution was on the order of 1 cm⁻¹. Particles within each mosaic, constructed using the LabSpec6 (LS6) SmartView configuration, were analyzed using the Particle Finder application V2. LS6 SmartView determines the topography $(\pm 50 \,\mu\text{m})$ and saves the focal point of all particles on the captured micrograph, enabling the stage to rapidly move the relevant particle into focus. The micrograph is converted into an 8-bit 0-255 greyscale image in which parameters are set by the user to visually separate particles from the darker filter substrate. Each particle was analyzed for 1 s by 2 accumulations at the above-described settings.

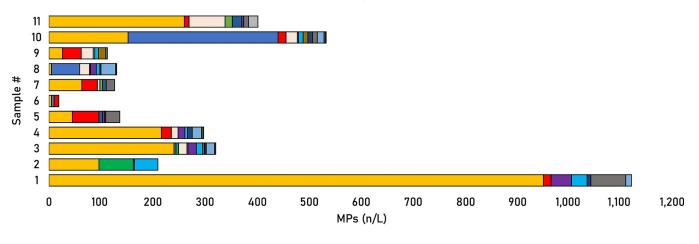
2.3. Spectral matching and verification

Using the Spectragryph spectral analysis software V1.2.17d (Dr. Friedrich Menges SoftwareEntwicklung, www.effemm2.de/spectragryph), all raw spectra were processed using adaptive baseline correction with 15% coarseness. The processed spectra were cross-referenced for their entire spectral range, using our in-house library containing selected spectra from the SLoPP and SLoPP-E [22] and the Cabernard [23] spectral libraries, also including self-obtained inhouse polymer spectra. Spectral matches were denominated by hit quality index (HQI)-values from 0 to 100% match. Spectra rated above 65% HQI were considered as MP candidates and were manually inspected and sorted by a trained interpreter to determine their validity. All morphological and chemical data, including Raman spectral data, is available in the <u>S1</u> and <u>S2 Datas</u>.

3. Results and discussion

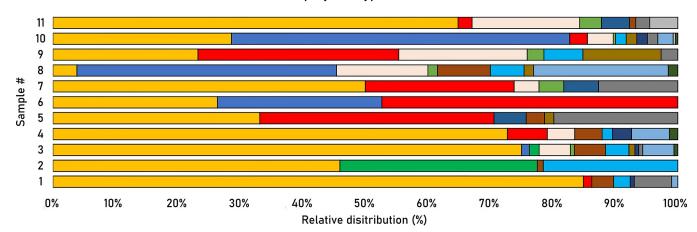
3.1. Microplastic concentration and polymer type distribution

MP concentrations in the ten investigated bottled water samples ranged from 19 to 1,154 MPs (n/L) following blank- and recovery corrections, demonstrating considerable variation with a mean concentration of 306 ± 316 n/L (Fig 1). These results suggest that the degree of MP



MP concentration and polymer type distribution

■PE ■PP ■PET ■PVC □PA6 ■PA6,6 ■PC ■PI ■PU ■PS ■CA ■PTFE ■PMMA ■PLA ■PBT



Relative polymer type distribution

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exposure through ingestion of bottled water is highly dependent on the specific brand of bottled water. However, in terms of numbers of MPs ingested, MP exposure may be several orders of magnitude lower than through indoor MP inhalation, even at the highest concentrations observed [24]. However, the toxicological impact of ingestion and inhalation of MPs may not be directly comparable [11, 25].

In total, n = 17 unique polymer types were identified. The most abundant polymer type detected was polyethylene (PE) which was identified in all samples in concentrations varying from 3.8 to 84.9%, followed by polypropylene (PP), polyethylene terephthalate (PET) and polyamide 6 (PA6). Although all investigated bottled water variants were stored in PET containers, PET was only detected in 7 out of 10 brands. Notably, in 3 of those 7 brands, the concentration of PET comprised less than 5% of identified MPs, demonstrating that the containers were not significant MP sources.

Fig 1. MP concentration (n/L) and relative polymer type distribution. Samples #1–10 present different, anonymous brands of bottled water. Sample #11 represents municipal tap water from Toulouse Metropole, France.

The MP concentration in municipal tap water from the Toulouse Metropole, France, was determined at 413 MPs (n/L) [0.096 μ g/L] and was higher than 8 out of 10 investigated bottled water brands. These relatively high concentrations may be attributed to the local water treatment process as tap water from this region is sourced by treating surface water from the Garonne river [26].

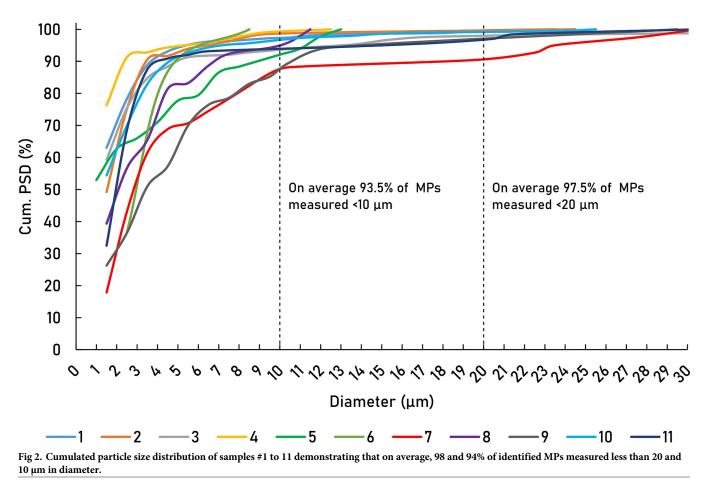
The MP concentration in potable water from the Toulouse Metropole area was approximately 10 times higher than concentrations reported in groundwater-sourced potable water in Denmark [39 n/L], which was also analyzed using Raman microspectroscopy [13]. Our findings are consistent with concentrations of MPs $\geq 1 \mu m$ detected in potable water from Czechish water treatment plants, ranging from 338 ± 76 to 628 ± 28 n/L, also analyzed using Raman microspectroscopy [14]. In Norway, potable water sourced from mountain lakes and processed at a water treatment plant was analyzed for MP concentration using pyrolysis gas chromatography-mass spectrometry (Py-GCMS) [27]. Their analysis demonstrated MP concentrations ranging from 0.006–0.093 μ g/L, similar to the current study's calculated values of tap water at 0.096 µg/L and bottled water ranging from 0.001 to 0.250 µg/L. A recent study on MP content in bottled water suggested much higher concentrations in terms of numbers of MPs $\geq 1 \,\mu m [24k \pm 13k n/L] [8]$, compared to the current study. however, the results of Qian et al. (2024) are ineligible for comparison as their study did not present a valid procedural blank control, meaning that their results are likely overestimated [28]. These results indicate that bottled water and treated surface water contain similar concentrations of MPs, while groundwater-sourced drinking water may be less contaminated. Different brands of bottled water, however, exhibited significant variation in microplastic (MP) concentrations, ranging up to two orders of magnitude [19 to 1,154 MPs/L]. Some brands were as clean as, or even cleaner than, groundwater-sourced potable water.

3.2. Microplastic size distribution

The EU directive 2020/2184 accepts a limit of detection of 20 μ m and imposes that at least 1,000 L of water should be filtered and analyzed per sample [10]. The current study demonstrated that 4.5 L of potable water was sufficient to reliably determine the MP concentration and polymer type distribution of MPs from 1–20 μ m above the contamination background. While a higher volume of sample generally provides better representation of the analyte in question, excessive quantities can lead to particle agglomeration [29]. Furthermore, our results demonstrated that 98% of all detected MPs measured less than 20 μ m in diameter (Fig 2). The largest identified MPs in the individual samples, ranged from 9 to 58 μ m in diameter and in 3 out of 11 samples, MPs above 20 μ m were not detected.

On average 94% of all detected MPs measured less than 10 μ m in diameter, demonstrating the prevalence of fine MPs, which are considered to have the highest implication for human health [11]. These results are in close agreement with [14] who determined that 95% of all detected MPs measured less than 10 μ m in diameter in treated drinking water.

Due to the presumed toxicological impact of nanoplastics (NPs), these should also be assessed [30]. However, when opting for a particle-based assessment by application of vibrational microspectroscopy techniques, NP assessment is currently commercially unfeasible [31]. Until routine particle-based NP detection becomes possible, the focus should be on identifying MPs from 1 μ m, while exploring methods such as alignment analysis to estimate NP concentrations [32]. Otherwise, absolute NP mass can be determined by mass spectrometry-based analytical methods [27, 33] but does not provide information on particle size distribution.



Cumulated particle size distribution

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4. Perspectives

4.1. Microplastic toxicity in humans

Plastic additives, some of which are known for their endocrine disrupting features [34], and other severe health effects [35–37], make up 6 wt.% of all plastic production [38]. In addition, plastic production workers and inhabitants adjacent to plastic production and waste disposal sites experience increased risk of premature birth, low birth weight, asthma, childhood leukemia, cardiovascular disease, chronic obstructive pulmonary disease, and lung cancer [35]. In addition to chemical toxicity, it has been suggested that also the inert nature of synthetic polymers can trigger oxidative stress and inflammation [39, 40] as well as carcinogenic responses [41].

The World Health Organization (WHO) considers that MPs <10 μ m have the highest implication for human health [11] due to their ability to penetrate into organ tissue [42]. Conversely, MPs >150 μ m are unlikely to be absorbed by the intestines [43]. In addition, plastics also have the ability to act as vectors for persistent organic pollutants [44]. In vitro toxicological experiments using polystyrene MP beads have demonstrated hepatotoxicity, being negatively size dependent and positively concentration dependent [45, 46]. However, inducing ecotoxicological responses typically requires exposure levels significantly higher than those found in the

environment. For instance, rodent studies introduced MP concentrations orders of magnitude higher than those observed in natural soils [47]. Nevertheless, recent medical research has shown that patients with carotid artery plaque containing MPs, faced elevated risks of myocardial infarction, stroke, or mortality [48], suggesting potential health implications of daily MP exposure.

Because MP research is a recent field of study, long-term and transgenerational health studies have yet to be completed, resulting in a knowledge gap in human health risks [49]. Yet, it has already been shown that plastic leachates can cause reproductive disruption [50], potentially linked to the world's decreasing fertility rates [51]. Therefore, similarly to pesticide regulations in potable water instated in the EU in 1998 [52], it might be preferable to appoint a low but feasible MP concentration threshold rather than abstaining from action altogether, even if the toxicological profile of MPs is not yet fully developed. For these reasons, it is important that the 1–20 μ m fraction of MPs is included in the assessment of plastic contamination in potable water.

5. Quality control and quality assurance

5.1. Negative control (blank correction)

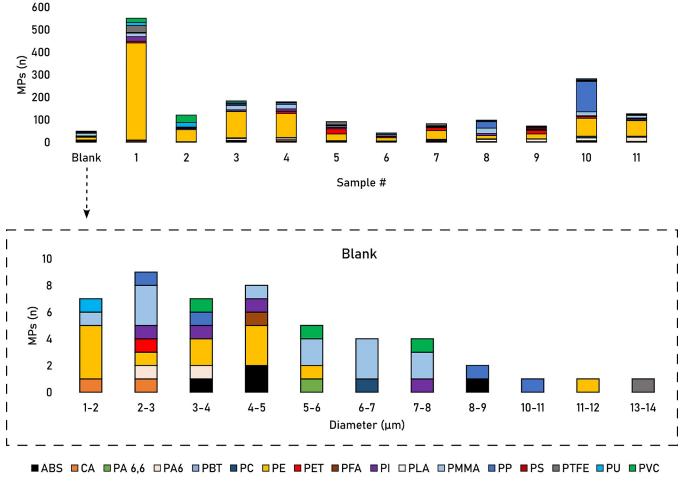
4.5 L of ultrapure grade-A milliQ water was filtered through 0.2 μ m, 47 mm aluminium oxide filter membranes (AnoDisc, Whatman), into individual kiln sterilized 1 L glass bottles, to remove potential MP contaminants, producing a MP-free solution to be used in the procedural blank experiment. The blank was treated according to the same protocol as the true samples to estimate and correct for procedural contamination during sample pre-treatment. During the analysis of three 2x2 mm grid subsamples, a total of n = 73,660 individual particles were analyzed by Raman microspectroscopy (mostly consisting of mineral residue from the kiln sterilized bottles), of which n = 49 particles were identified as MPs (Fig 3).

For each individual sample, a blank correction was performed on the basis of polymer type and particle size. When two particles in the primary sample and the blank matched in polymer type and size, they formed a 'pair'. Once a pair was identified, the MP particle in the primary sample was subtracted, and its matching particle in the blank could no longer be used for sub-traction. To form a pair, particles had to be of the same polymer type and the particle of the primary sample could not measure more than twice the diameter of the largest MP particle identified in the blank. Blank correction could be performed prior to extrapolation because similar fractions of filter area, corresponding to 7.8%, had been examined in all samples (with the exception of sample #11 at 3.9%).

5.2. Positive control (recovery correction)

To correct for unintentional MP loss during sample pre-treatment [53, 54], a procedural recovery experiment using a precise number of red polyethylene (PE) fragments in the 5–100 μ m range (EasyMP, Microplastic Solution, France), was conducted. In four individual samples of 4.5 L of grade-A milliQ water, a total of n = 1,074, 1,176, 1,112 and 1,086 MP fragments were intentionally added and the four samples were treated according to the same protocol as the true samples. Following the full protocol, the remaining number of spiked MPs was evaluated. Here a total of n = 1,056, 917, 645 and 874 MPs were recovered. The use of fragments instead of MP beads is not only more environmentally relevant [55] but also allows for the determination of analytical recovery within size groups [29] (Fig 4).

The recovery experiment demonstrated an increase in analytical MP recovery with decreasing size, leading to positive recovery (higher output than input) in the 5–15 μ m size range. We speculate that this positive analytical recovery is due to ultrasonication, causing MP



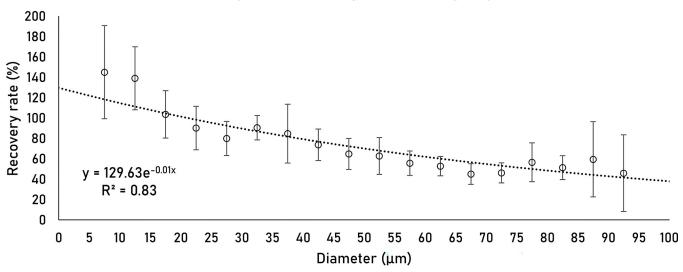
Total number of MPs identified within three 2x2 mm grid subsamples

Fig 3. In the three procedural blank subsamples a total of n = 49 MPs were identified. A diverse range of polymer types within the procedural blank suggests atmospheric input as the likely source of contamination, as no specific polymer type was dominant.

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fragmentation and particle breakup [56]. This notion conforms with observations of polyvinylidene fluoride (PVDF) micro-fragments found in all samples in high concentrations, including in the blank. During the protocol, PVDF filter membranes were used for filtration and incident particle transfer into the oxidant for MP isolation. To verify whether the observed PVDF microparticles resulted from fragmentation of the filter membrane, a new experiment was staged. Here, 4.5 L of bottled water from the same 6-pack of 1.5 L bottles that revealed the highest concentration of PVDF particles (sample # 4), was once again treated and analyzed. The sample was treated according to the same protocol as all the other samples, with the exception that the PVDF filter membrane had been replaced with a 0.2 μ m, 47 mm nitrocellulose membrane. In this experiment no PVDF particles were identified and it was therefore concluded that the protocol had caused fragmentation of the PVDF filter membrane, the red PE fragments, and incidentally the native MPs within the sample. From this experiment an exponential function (R² = 0.83) to describe the relation between particle size and analytical recovery rate (RR) was established (Eq 1).

$$RR(\%) = 129.63e^{-0.01 \cdot Diameter(\mu m)}$$
(1)



Mean microplastic recovery within size groups (%)

Fig 4. Analytical recovery of MPs within size groups, demonstrating a negative correlation between recovery rate and particle size. In the 5–15 μm size range, the analytical recovery showed positive values, possibly due to particle breakup caused by ultrasonication during sample pre-treatment.

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Eq 1 was applied to correct for the analytical recovery of all detected MPs within size groups on the order of 1 μ m.

5.3. Correction for unintentionally partitioned MP particles (merging tool)

Due to the frame-by-frame acquisition process necessary for obtaining accurate Raman measurements of MPs down to 1 μ m in diameter, particles located on the edge of a given frame (edge-particles) are incidentally partitioned and identified as multiple particles. At the current microscopic resolution, a single frame measures 60x40 μ m. Consequently, this leads to an overestimation of the smaller size fraction and underestimation of larger particles. The LS6 Particle Finder application offers a built-in option to ignore edge particles; however, this approach can result in underestimating the true MP concentration. It disproportionately excludes larger particles, which are more likely to intersect with the edge of the frame.

Therefore, a custom script (Microplastic Solution, France) was developed to improve postprocessing of data by merging unintentionally partitioned MPs. Due to commercial interests, the code cannot be shared but the principal functions are disclosed. The script measures the distance between all particles and determines if particles of the same polymer type are overlapping. 'Overlap' is defined as the distance between the center of two particles being smaller than the major axis of one the particles in question. If multiple particles of the same polymer type are overlapping, these are grouped and the particle with the highest spectral hit quality index (HQI) is defined as the 'leader' of the group of which its index number is assigned to define the new, merged particle. The merged particle attains the cumulative area of the group and the remaining particles within the group are discarded. Similar to how diameter is calculated in the LS6 Particle finder application, the diameter of the merged particle is calculated from the observed area by the assumption of a circular model *i.e.* area-equivalent diameter (Eq 2).

$$D = 2 \cdot \sqrt{\frac{A}{\pi}} \tag{2}$$

Minor and major axes are observed values and must thus be recalculated for the merged

particle based on the average deviation from diameter, determined at $+40\% \pm 17\%$ and $-30 \pm 13\%$ for major and minor axes, respectively. These estimates were determined based on the average deviation between diameter and the two parameters, calculated from a dataset consisting of more than 5,000 identified MPs from a previous project on atmospheric MP deposition, where edge-particles had been excluded.

6. Study limitations and future prospects

6.1. Repeatability

To obtain MP concentration estimates of sufficient accuracy and credibility, three replicates each of at least 5 L of sample should be treated and assessed on a membrane measuring between 10 to 25 mm in diameter. In each replicate, a single 2x2 mm grid should be examined in the center of the filter to assess the 1–20 μ m fraction. The 1–20 μ m fraction should be examined by application of Raman or O-PTIR [57] microspectroscopy. Sufficient MPs should be subsampled (n \geq 96) to meet statistical significance [58].

With the aim of examining a higher number of particles, analyzing a larger area compensates for processing more sample and is often more comprehensive. For example, filtering X volume of sample and analyzing X μ m² is preferable to filtering 2·X volume of sample and analyzing X/2 μ m² of filter surface. In theory both approaches would lead to the same number of particles appearing on filter membrane. However, a greater spread between individual particles reduces the likelihood of agglomeration, which is preferable when performing vibrational microspectroscopic analysis [29].

6.2. Protocol adjustments

It is likely that the ultrasonication step, intended to enhance particle transfer from the filter membrane to the oxidant during sample pre-treatment, caused the breakup of both intrinsic MPs and the filter membrane itself. If particle breakup can be avoided while maintaining acceptable analytical recovery, ultrasonication should be omitted from the protocol.

7. Conclusions

Ten bottled water samples of different brands were investigated for microplastic (MP) content by Raman microspectroscopy, revealing concentrations ranging from 19 to 1,154 MPs (n/L) following blank- and recovery corrections, demonstrating considerable variation with a mean concentration of 306 ± 316 n/L. These results suggest that the degree of MP exposure through ingestion of bottled water is highly dependent on the specific brand of bottled water. The MP concentration in municipal tap water from the Toulouse Metropole, France, was determined at 413 MPs (n/L) [0.096 µg/L] and was higher than 8 out of 10 investigated bottled water brands. The MP concentration was in close resemblance with other sources of treated surface drinking water in Norway and the Czech Republic but was ~10 times higher than groundwater-sourced drinking water in Denmark. On average, 98 and 94% of detected MPs measured less than 20 and 10 µm in diameter, demonstrating the prevalence of fine MPs in potable water.

In total, n = 17 unique polymer types were identified from which polyethylene (PE), polypropylene (PP), polyethylene terephthalate (PET) and polyamide 6 (PA6) made up the majority of identified MPs. Although all bottled water variants were stored in PET containers, PET was only present in 7 out of 10 bottled water brands. In 3 of those 7 brands, PET comprised less than 5% of identified MPs, demonstrating that the containers were not significant MP sources. By demonstrating that the comprehensive analysis of MPs from to $1-20 \mu m$ in potable water is feasible, and that this fraction makes up 98% of all detected MPs, likely posing the greatest risk to human health, we strongly suggest that the detection of the $1-20 \mu m$ fraction be incorporated into regulatory guidelines for assessing plastic contamination in potable water, including the EU Directive 2020/2184. The current protocol serves as a viable method to detect MPs of any size in any type of potable water regardless of hardness.

Supporting information

S1 Data. Contains morphological information and chemical identity of all examined particles in all samples. (XLSX)

(ALSA)

S2 Data. Contains Raman spectral data (baseline corrected) of all examined particles in all samples, in.sgd (SpectraGryph spectra file) format. The data file can be downloaded from the Dryad repository: doi.org/10.5061/dryad.z612jm6nb. (TXT)

Author Contributions

Conceptualization: Oskar Hagelskjær, Henar Margenat, Nadiia Yakovenko.

Data curation: Oskar Hagelskjær.

Formal analysis: Oskar Hagelskjær.

Funding acquisition: Jereon E. Sonke, Gaël Le Roux.

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Methodology: Oskar Hagelskjær, Nadiia Yakovenko.

Project administration: Oskar Hagelskjær.

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Supervision: Jereon E. Sonke, Gaël Le Roux.

Validation: Jereon E. Sonke, Gaël Le Roux.

Visualization: Oskar Hagelskjær.

Writing - original draft: Oskar Hagelskjær, Jereon E. Sonke.

Writing - review & editing: Oskar Hagelskjær.

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