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 bottled water can 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, small MPs in the 1-20 µm range are far more likely to pass the human intestine into blood and organs. We therefore investigated MP concentrations down to 1 µm in ten individual polyethylene terephthalate (PET) bottled water brands and one tap water sample by Raman microspectroscopy. Our analyses are supported by procedural blank- (negative control) and analytical recovery correction (positive control) using red polyethylene fragments in the 5-100 µm range. We find that MP concentrations range from 19 to 1,154 (n/L) [0.001 to 0.250 µg/L]. 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.

Keywords: drinking water, EU directive 2020/2184, pollution, health, methodology, 1-20 µm fraction.

1. Introduction

A ‘Microplastic’ (MP) particles is defined as any plastic item which longest axis measures between 1 to 5000 µm (5 mm) (Barnes et al., 2009; Lassen et al., 2015) or 1 to 1000 µm (1 mm) according to the International Organization for Standardization (ISO, 2023). MPs are ubiquitous pollutants (Hale et al., 2020) and have inevitably made their way into our nutrition (Mamun et al., 2023) and potable water supply; both tap and bottled (Gambino et al., 2022). Bottled water especially, has a bad reputation in terms of MP

contamination (Mason et al., 2018; Qian et al., 2024) and on a global, annual base we consume more than 60 L of bottled water per capita (Statista, 2024).

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 (Mason et al., 2018), 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 (EU, 2024). The directive accepts a limit of detection of 20 µm despite MPs <10 µm being considered most relevant to human health (WHO, 2022). In addition, MPs <10 µm are significantly more abundant in potable water (Maurizi et al., 2023) and therefore it is crucial to encompass the entire size spectrum of MPs (1-5000 µm) for a comprehensive assessment of MP contamination in any product intended for human consumption.

However, prior to enacting potential regulations, it is important that we establish our ability to comprehensively measure and classify MPs down to 1 µm in potable water. To determine the polymeric composition of MPs from 1 µm to 5 mm, we employed Raman microspectroscopy in combination with a computer vision based post-processing software (microplasticsolution.com, France) used to correct for unintentional particle partitioning. Between ten anonymous, French retail polyethylene terephthalate (PET) bottled water brands, more than half a million individual particles were investigated of which n = 1,824 were identified as synthetic polymers. We find concentrations 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.

2. Methods and materials

2.1. Laboratory pre-treatment

In ten different French, anonymous brands of bottled water, the concentration of MPs ≥1 µm was established by automated Raman microspectroscopy. Additionally, one sample of potable tap water from the Toulouse Metropole area was examined. For each sample, 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. Each filter membrane was transferred into 50 mL glass vials filled with 30 mL of 30 vol.% hydrogen peroxide (H_2O_2) (Fischer Scientific, Belgium). To improve the transfer from the filter membrane to the H_2O_2-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 H_2O_2 (30 vol.%) to impede particles from sticking onto the filter membrane thus increasing MP recovery. H_2O_2 (30 vol.%) The filter was flushed until the vial held 40 mL.

Following six continuous days of hotplate-induced digestion at 50°C, 5 mL of 5 vol.% hydrochloric acid (HCl) was added to each vial under a fume-hood, to increase the acidity of the solution and improve the digestion efficiency of carbonate minerals. The reagents together lead to a redox reaction that is weaker than Fenton's reaction, slowly forming hypochlorous acid (HOCl) and water (H_2O) (Yu et al., 2019). Following 24 h of reaction, each solution was filtered through individual 0.2 µm, 25 mm aluminium oxide filter membranes (Whatman Anodisc, U.K.) and flushed with ultrapure grade-A milliQ water (18.2 MΩ·cm), leaving the desired particles on a flat surface suitable for microspectroscopic Raman analysis (Hagelskjær et al., 2023b). 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. All sample manipulation was carried out inside a FlowFAST® V12P laminar flow cabinet within a dedicated microplastic laboratory. In addition, red cotton lab coats were worn at all times inside the laboratory.

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 blank. All particles measuring ≥1 µm in diameter were subject to Raman analysis. Raman measurements were carried out at 20°C using a Horiba (Jobin Yvon, France) LabRAM Soleil. 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 µm 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 LS6 Particle Finder application V2. LS6 SmartView determines the topography (± 50 µm) and saves the focal point of all particles on the captured photomicrograph, enabling the stage to rapidly move the relevant particle into focus. The photomicrograph 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.

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 (Munno et al., 2020) and the Cabernard (Cabernard et al., 2018) spectral libraries, also including self-obtained in-house 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.

3. Results and discussion

3.1. Microplastic concentration and polymer type distribution

MP concentrations in the eleven investigated samples ranged from 19 to 1,154 MPs (n/L). In total, n = 17 unique polymer types were identified (Fig. 1). Polyethylene (PE) was detected in all samples and concentrations varied from 3.8 to 84.9%, followed by polypropylene (PP), polyethylene terephthalate (PET) and polyamide 6 (PA6). High-pressure filtration membranes are often used in the treatment of potable water (ultrafiltration) and sometimes even fraudulently in contradiction with national regulations (Fourcart, 2024). These membranes can be manufactured in PE and PP (Boriskina, 2019; Ghodake et al., 2020), amongst other synthetic polymers including polytetrafluoroethylene (PTFE), polyvinylidene fluoride (PVDF), PA6 and PA6,6, polyacrylonitrile (PAN), etc. Due to an operating pressure difference between the entrance and the exit of the filtration membrane of 8 bar (Li et al., 2018), the resulting friction between filtration components and the potable water itself is a potential source of plastic fragmentation and incident MP generation, despite the initial intention of removing particulate contaminants.

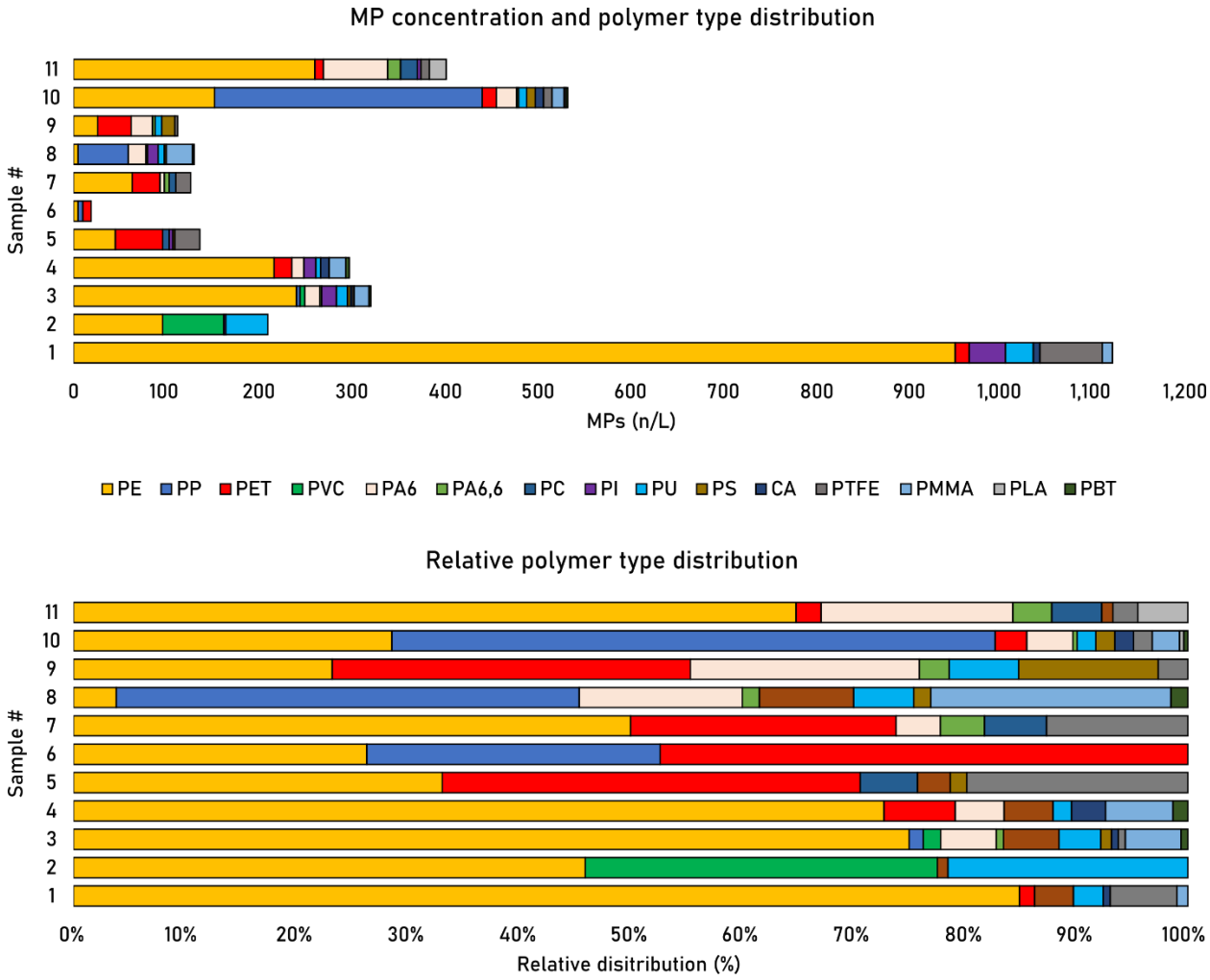

Figure 1 - MP concentration (n/L) and relative polymer type distribution. Samples #1-10 present different, anonymous brands of bottled water. Sample #11 represents municipal tap water from Toulouse Metropole, France.

PET, the polymer type from which all the investigated water bottles in the current study were constituted, was detected in 7 out of 10 brands. In 3 of those 7 brands, the concentration of PET comprised less than 5% of identified synthetic polymers, demonstrating that the container itself is not necessarily an important source of MP contamination. The MP concentration in municipal tap water from the Toulouse Metropole, France, was higher than 8 out of 10 investigated bottled water brands. These relatively high concentrations are likely attributed to the local water treatment process as tap water from this region is primarily sourced by treating surface water from the Garonne river (Toulouse Metropole, 2024). MP concentrations were estimated at 413 MPs (n/L) [0.096 µg/L] which in terms of mass is 35 times higher than concentrations reported in groundwater-sourced potable water in Denmark [0.0028 µg/L] (Maurizi et al., 2023), where ultrafiltration is not applied. This evidence supports the notion that ultrafiltration of potable water may lead to MP contamination in potable water. Recent research on MP in bottled water suggested much higher

concentrations in terms of numbers of particles (Qian et al., 2024), however, these results cannot be used for comparison as no procedural blank control in Qian's study was conducted.

3.2. Microplastic size distribution

The EU directive 2020/2184 accepts a limit of detection of 20 µm and imposes that at least 1,000 L of water should be filtered and analyzed per sample (EU, 2024). 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 µm above the contamination background. While it is true that a higher volume of sample generally provides better representation of the analyte in question, excessive quantities can lead to particle agglomeration (Hagelskjær et al., 2023a). Furthermore, our results demonstrated that 98% of all detected MPs measured less than 20 µm in diameter (Fig. 2). The largest identified MPs within individual samples, ranged from 9 to 58 µm in diameter and in 3 out of 11 samples MPs above 20 µm were not detected. These results suggest that the exposure to MPs >20 µm through consumption of certain brands of bottled water is very low, considering that the average person ingests 2 L of potable water per day (Westrell et al., 2006). In addition, exposure to MPs through ingestion of certain bottled water brands may be several orders of magnitude lower than through daily indoor inhalation (Eberhard et al., 2024).

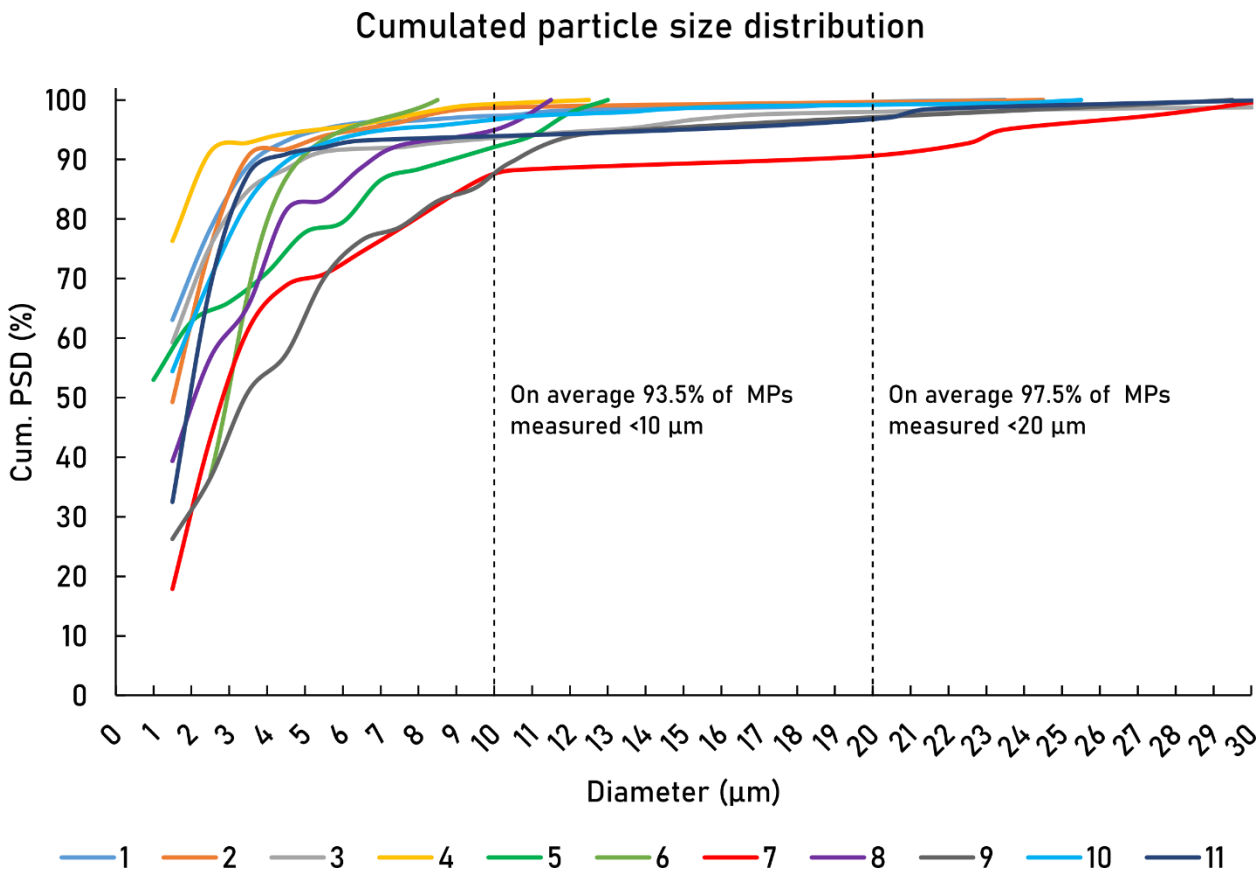

Figure 2 - Cumulated particle size distribution of samples #1 to 11 demonstrating that on average, 98 and 94% of identified MPs measured less than 20 and 10 µm in diameter.

On average 94% of all detected MPs measured less than 10 µm in diameter, demonstrating the prevalence of very small MPs which are considered by the World Health Organization (WHO) to have the highest implications for human health (WHO, 2022). Due to their toxicological impact, nanoplastics (NPs) should also be assessed (Busch et al., 2023). However, when opting for a particle-based assessment by application of vibrational microspectroscopy techniques, NP assessment is currently commercially unfeasible (Shorny et al., 2023). Until particle-based NP detection becomes feasible, our focus should be on identifying MPs from 1 µm, while exploring methods such as alignment analysis to estimate NP concentrations (Koelmans et al., 2020). Otherwise, absolute NP mass can be determined using mass spectrometry-based analytical methods (Velimirovic et al., 2021) but does not provide information on the particle size distribution.

A correlation between the average diameter of all identified MPs in each individual sample was plotted against MP concentration (n/L), illustrating a negative correlation between particle size and MP concentration (Fig. 3). This indicates that as concentrations rise, MP particles tend to decrease in size. Perhaps, a high concentration of small MPs could stem from the fragmentation of fibrous filtration

membranes. This occurrence could be attributed to the release of exposed extruding members from newly installed membranes or, conversely, from older deteriorating filter membranes. Other plastic contact-points within treatment facilities, such as pipes and containers also warrant consideration but none of these components exhibit a comparable surface-area-to-volume ratio under conditions of high pressure.

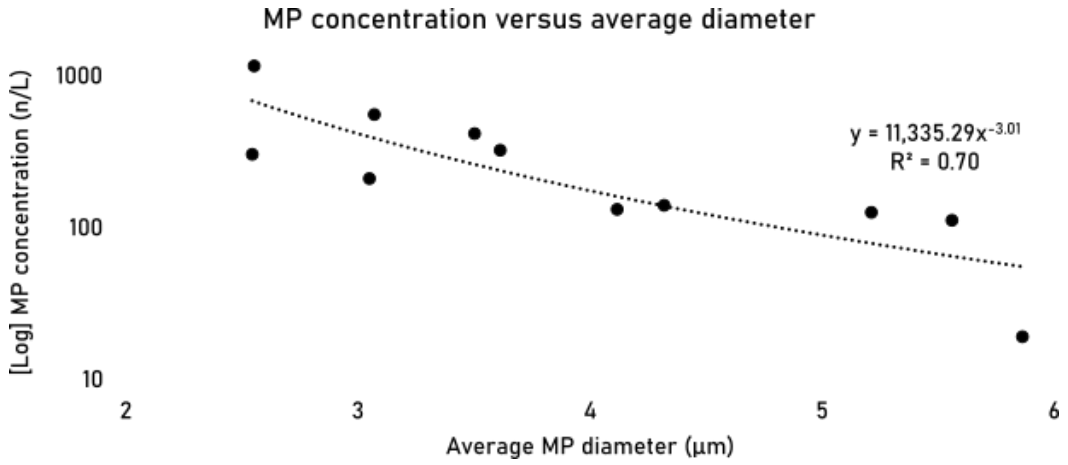

Figure 3 - Average MP diameter plotted against MP concentration in all investigated samples, demonstrating a negative correlation between the two parameters. Labels provide sample numbers.

4. Perspectives

4.1. Microplastic toxicity in humans

Plastic additives, some of which are known for their endocrine disrupting features (Quignot et al., 2012), and other severe health effects (Landrigan et al., 2023; Maddela et al., 2023; Trasande et al., 2024), make up 6 wt.% of all plastic production (Geyer, 2020). 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 (Landrigan et al., 2023). In addition to chemical toxicity, it has been suggested that also the inert nature of synthetic polymers can trigger oxidative stress and inflammation (Büks et al., 2020; Yoon et al., 2021) as well as carcinogenic responses (Choudhury et al., 2023). The World Health Organization (WHO) considers that MPs <10 µm have the highest implication for human health (WHO, 2022) due to their ability to penetrate into organ tissue (Kadac-Czapska et al., 2023). Conversely, MPs >150 µm are unlikely to be absorbed by the intestines (EFSA, 2016). In addition, plastics also have the ability to act as vectors for persistent organic pollutants (Saud et al., 2023). In vitro toxicological experiments using polystyrene MP beads have demonstrated hepatotoxicity, being both size- and concentration-dependent (Cheng et al., 2022; Hua et al., 2022). 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 (Mills et al., 2023). Nevertheless, recent medical research has shown that patients with carotid artery plaque containing MPs, faced elevated risks of myocardial infarction, stroke, or mortality (Marfella et al., 2024), 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 (Kirstein et al., 2021). However, it has already been shown that plastic leachates can cause reproductive disruption (Akoueson et al., 2023), potentially linked to the world's decreasing fertility rates (Aitken, 2024). We also know that adjacency to plastic production or discarding-sites can have serious health effects including cancer (Landrigan et al., 2023). Therefore, similarly to pesticide regulations in potable water instated in the EU in 1998 (Dolan et al., 2013), 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 µm 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 µm, 47 mm aluminium oxide filter membranes (AnoDisc, Whatman) into individual kiln sterilized 1 L glass bottles, to remove potential MP contaminants, producing a microplastic-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 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. 4).

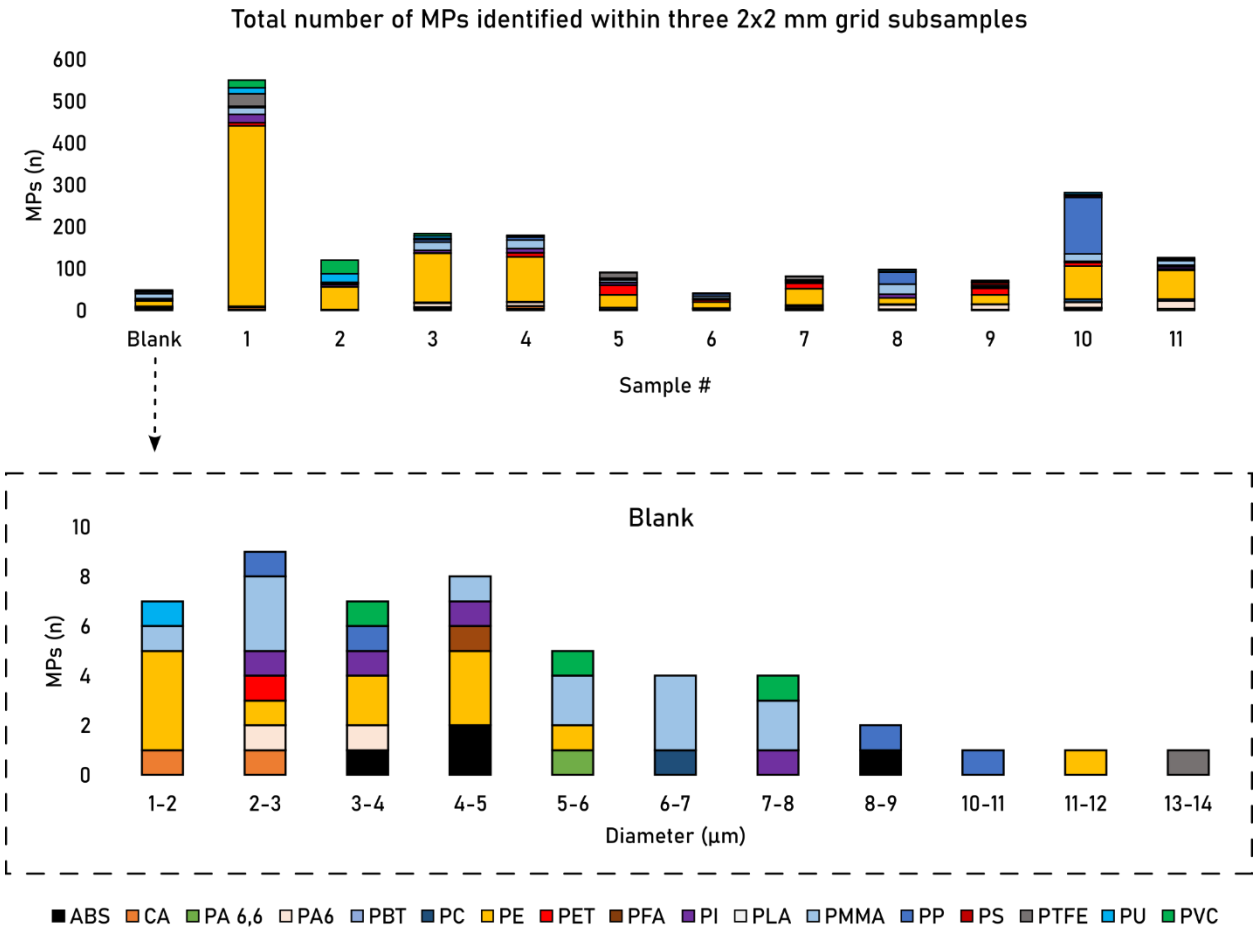

Figure 4 - In the three procedural blank subsamples a total of n = 49 MPs were identified. A diverse range of polymer types within the procedural blank suggests atmospheric input as the likely source of contamination, as no specific polymer type was overrepresented.

For each individual sample, a blank correction was performed on the basis of polymer type and particle size. If a MP particle of a specific polymeric type was identified in both the blank and the true sample, the MP particle of that specific polymeric within the nearest size range was removed from the true sample prior to extrapolation. 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 subtraction. 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 particle loss during sample treatment (Dimante-Deimantovica et al., 2022; Way et al., 2022), a procedural recovery experiment using a precise number of red polyethylene (PE) fragments in the 5-100 µm range (PrecisionMP™, microplasticolution.com, 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 (Rozman and Kalčíková, 2022) but also allows for the determination of analytical recovery within size groups (Hagelskjær et al., 2023a) (Fig. 5).

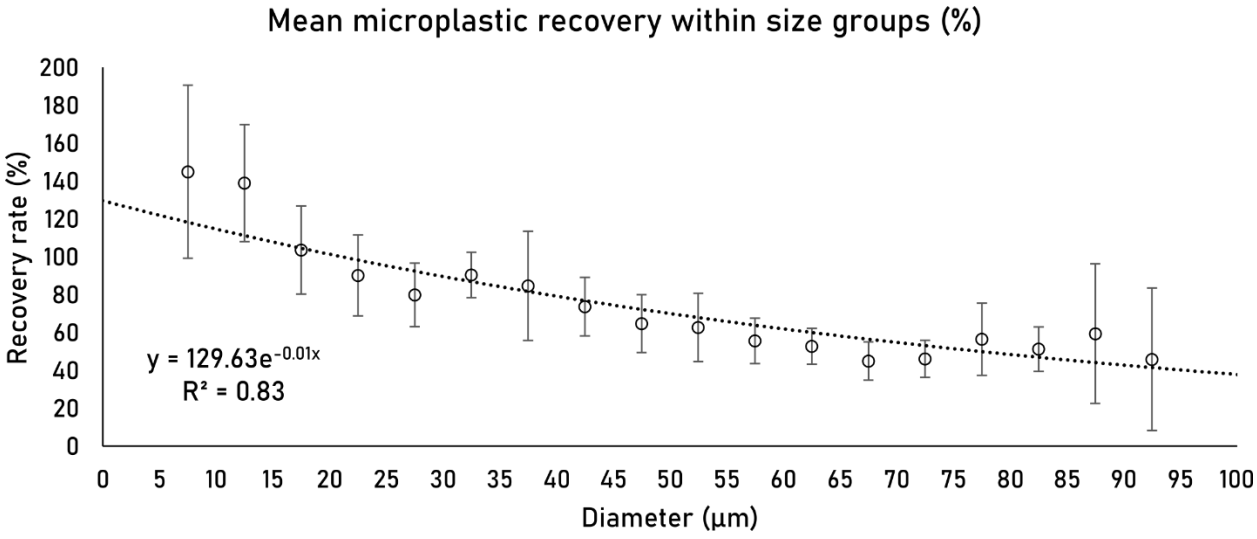

Figure 5 - Analytical recovery of MPs within size groups, demonstrating a negative correlation between recovery rate and particle size. In the size range of 5-15 µm, the analytical recovery even reached positive values, possibly explained by particle breakup due to ultrasonication during sample pretreatment.

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 µm size range. We speculate that this positive analytical recovery is due to ultrasonication, causing MP fragmentation and particle breakup (Vollertsen and Hansen, 2017). This notion conforms with observations of polyvinylidene fluoride (PVDF) microparticles 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. 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 by a 0.2 µm, 47 mm nitrocellulose membrane. The experiment demonstrated that no PVDF particles could be identified and we could therefore conclude that the protocol did indeed cause fragmentation of the PVDF filter membrane, and most likely, of the spiked red PE fragments used in the recovery experiment. Incidentally, an exponential function (R^2 = 0.83) to describe the relation between particle size and analytical recovery rate (RR) was established (Eq. 1).

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

Because we assume that the intrinsic MPs within each true sample responded similarly to the protocol, Eq. 1 was applied to all detected MPs to correct for the analytical recovery within size groups on the order of 1 µm.

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 µm in diameter, particles located on the edge of a frame (edge-particles) are incidentally partitioned and identified as multiple particles. At the current microscopic resolution, a single frame measures 60x40 µm. Consequently, this leads to an overestimation of the smaller size fraction and underestimation of larger particles. Although the software has a built-in option to ignore edge-particles, this would also lead to underestimation of the true MP count and would favor exclusion of large particles, as these are more likely to come in contact with the edge of the frame.

Therefore, a custom script (microplasticsolution.com, France) was developed to improve post-processing of data by merging unintentionally partitioned MPs. Due to commercial interests, the script cannot be shared but the principal functions are disclosed. The script measures the distance between all particles and determines if any 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 (Eq. 2) [area-equivalent diameter].

$$D = 2 \cdot \sqrt{\frac{A}{\pi}} \quad (2)$$

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 *positive* 40% ± 17% and *negative* 30 ± 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 higher accuracy and credibility, three replicates each of at least 5 L of sample should be treated and assessed on a membrane measuring between 13 to 25 mm. In each replicate, a single 2x2 mm grid should be examined in the center of the filter to assess the 1-20 µm fraction. The 1-20 µm fraction should be assessed using Raman or O-PTIR (Böke et al., 2022) microspectroscopy. If these criteria are met, enough MPs should be subsampled ($n \geq 96$) to meet statistical significance of the established MP concentration (Cowger et al., 2024), unless the sample is clean. With the aim of examining a higher number of MPs, analyzing a larger area compensates for processing less sample and is often more comprehensive. For example, filtering X volume of sample and analyzing X μm^2 is preferable to filtering $2 \cdot X$ volume of sample and analyzing X/2 μm^2 of filter surface. In theory both approaches would lead to the same number of particles on the investigated area but with the first approach there would be greater spread between individual particles and less likelihood of agglomeration (Hagelskjær et al., 2023a), which is preferable when performing vibrational microspectroscopy.

6.2. Protocol adjustments

Most likely, the ultrasonication step intended to improve particle transfer from the filter membrane to the oxidant during the sample pre-treatment, caused particle breakup of the intrinsic MPs but also of the filter membrane itself. As a consequence, the recovery experiment should be repeated without the application of ultrasonication to determine whether this step leads to particle breakup. If particle breakup is reduced and analytical recovery is acceptable, ultrasonication should be dismissed from the protocol. Alternatively, pre-sonication of the membrane before filtration can be explored to limit its fragmentation during a 2nd sonication step.

7. Conclusions

The microplastic (MP) concentration in ten individual, French bottled water brands and one tap water sample was evaluated by application of Raman microspectroscopy. MP concentrations in the eleven investigated samples ranged from 19 to 1,154 MPs (n/L) [0.001 to 0.250 µg/L]. In total, n = 17 unique

polymer types were identified from which PE, PP, PET and PA6 made up the majority of identified MPs. PET was only present in 7 out 10 bottled water brands and in 3 of those 7 brands, the concentration of PET comprised less than 5% of identified synthetic polymers, demonstrating that the container itself is not necessarily an important source of MP contamination. Average MP particle diameter was lower in samples with higher MP concentrations and we suspect that one of the primary MP contamination sources in potable water is linked to ultrafiltration and incident fragmentation. 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. For individual brands, the largest identified MPs measured between 9 and 58 µm in diameter. Laboratory contamination was corrected for by application of a procedural blank experiment (negative control) using pre-filtered [0.2 µm] MilliQ water in the same volume as the true samples. Blank correction was performed on a particle-to-particle basis. Analytical recovery (positive control) was evaluated through the execution of a MP recovery experiment, using red polyethylene fragments in the 5-100 µm range. The experiment revealed a negative correlation between particle size and recovery rate, where particles from 5-15 µm demonstrated positive recovery (larger output than input) which we interpret as particle breakup during sample treatment. This notion was further supported by the evidence of fragmentation of the polyvinylidene fluoride (PVDF) filter membrane used for particle transfer. By demonstrating that the comprehensive analysis of MPs down to 1 µm in potable water is feasible using relatively small volumes of water (~5 L/sample), and by considering the most recent perspectives in MP risk assessment for human health, we strongly suggest that the 1-20 µm fraction of MPs should be included in the assessment of plastic contamination in potable water.

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9. Declarations

9.1. Acknowledgements

Not available

9.2. Authors' contribution

O.H. conceptualized and administered the project, led the laboratorial work, produced and interpreted data and led manuscript writing. F.H. developed and wrote the script used to improve data post-processing. H.M and N.Y. assisted in maintenance of the Raman instrument and provided criticism to the project and manuscript. J.E.S and G.L.R. secured the funding, supervised the project and provided critical revision of the manuscript.

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9.4. Competing interests

The authors declare no conflict of interest.

9.5. Availability of data and materials

All data procured during this study are included in the published article and its supplementary information files.

9.6. Abbreviations

MP (microplastic), NP (nanoplastic), PE (polyethylene), PP (polypropylene), PET (polyethylene terephthalate), PS (polystyrene), PA (polyamide), PVC (polyvinyl chloride), PU (polyurethane), ABS (acrylonitrile butadiene styrene), PMMA (polymethyl methacrylate), PC (polycarbonate), PBU (polybutadiene), PLA (polylactic acid), PFA (perfluoroalkoxy alkane), PI (polyisoprene), PTFE (polytetrafluoroethylene), PVDF (polyvinylidene fluoride), EU (European Union), FT-IR (Fourier transform infrared), µm (micrometer), mm (millimeter), mL (milliliter), L (liter), µg (microgram), n (number).

9.7. Ethics approval

Not applicable.

9.8. Consent for publication

Not applicable.

10. Figure captions

Figure 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.

Figure 2: Cumulated particle size distribution of samples #1 to 11 demonstrating that on average, 98 and 94% of identified MPs measured less than 20 and 10 µm in diameter.

Figure 3: Average MP diameter plotted against MP concentration in all investigated samples, demonstrating a negative correlation between the two parameters. Labels provide sample numbers.

Figure 4: In the procedural blank, 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 overrepresented.

Figure 5: Analytical recovery of MPs within size groups, demonstrating a negative correlation between recovery rate and particle size. In the size range of 5-15 µm, the analytical recovery even reached positive values, possibly explained by particle breakup due to ultrasonication during sample pretreatment.

11. Supplementary information captions

Supplementary information 1

12. Highlights

- 98 and 94% of all detected microplastics measured less than 20 and 10 µm in diameter.
- Polyethylene terephthalate (PET) is not necessarily an important source of microplastic contamination.
- Microplastic (>1 µm) concentrations in ten brands of bottled water diverged from 19 to 1,154 n/L.