Systematic Review

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Microplastics in water, from treatment process to drinking water: analytical methods and potential health effects

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Abstract

Aim: The commonly used analytical methods for microplastic (MPs) detection in drinking water and the threat of MP pollution in water intended for human consumption to human beings are presented through a systematic review. Furthermore, MP occurrence, transport, and fate from raw to treated drinking water, tap water, and bottled water, as well as the possible health impacts of MPs on human beings, are also evaluated.

Methods: Systematic review included articles published in scientific journals that contain specific keywords in the title and were searched in Web of Science (WOS) and Scopus. The literature was selected and extracted by two reviewers based on the PRISMA-A guidelines, which recommend including 57 items.

Results: The experimental studies pointed out that sampling is performed using grab or reduced samples, and sample treatment involves mostly oxidation with hydrogen peroxide and density separation. The minimum sample size obtainable in the extraction and the maximum density of the polymer separable from the matrix provided different results. Clearly, the determination of MPs involves the simultaneous application of several analytical

techniques, including optical, fluorescence, and electronic microscopies, µFTIR, µ-Raman, and pyrolysis gas chromatography-mass spectrometry. The determination technique also provides different results according to the sensitivity as well as the minimum size determinable. These studies are mostly devoted to establishing the occurrence, transport, and fate within the supply network, the efficiency in removal of MPs from drinking water by treatment plants, and the risk to humans. The MP concentration in drinking water reservoirs is highly variable. However, tap water always presents lower concentrations of MPs than the water that enters the drinking water treatment plants because the different treatments are efficient at removing MPs. Although it has not been fully demonstrated that MPs are toxic to humans, the effects point to oxidative stress, gastrointestinal irritation, microbiome irregularities, and changes in lipid metabolism.

Conclusion: Analytical methods present some common features as a first step towards harmonization. However, it is still unknown whether the analytical methods could influence the disparity of the results. The MP concentration in drinking water is low in comparison to other types of water. MPs are not exempt from hazards to human health.

Keywords: Plastics, raw water, tap water, bottled water, drinking water treatment plants, risk assessment

INTRODUCTION

Plastic production and usage have exponentially increased in the last few years, and it has become an essential part of our daily life. Data show that the annual global plastic production as of 2019 is more than 369 million tons a year[1], and it is estimated that it may exceed a billion tons by 2050[2,3]. Its widespread use and the fact that the complete biodegradability of plastic materials can take up to 450 years mean plastics tend to accumulate in different types of environments, especially in aquatic ecosystems where, if they were not already released as microplastics (MPs), they can result from the breakdown of plastics by physical or chemical erosion or due to the action of UV light either on land or in water[4,5]. MPs include a wide range of materials (thousands of different plastics), each with its own chemical composition and characteristics, such as size (ranging from 0.1 to 5000 µm), shape (fibers, films, pellets, fragments, and foams), and color (transparent, red, green, blue, black, *etc.*)[6].

In recent years, MPs have been extensively detected in seawater, freshwater, and wastewater[7-12]. A study in 2014 already revealed that there are more than five trillion pieces of plastic floating on the surface of the world seas[13]. Several studies have highlighted that recent events such as the COVID-19 pandemic exponentially increased the presence of MPs in coastal areas and other water bodies[14,15]. In this context, some studies have also demonstrated that the plastic pollution in drinking water is as serious as that in the seas. Liu *et al.*[11], after reviewing 53 studies, established the median MP concentration in conventional water sources as 2.2×10^3 items m^{-3}, with the size of particles identified usually > 50 µm. Similarly, Cheng *et al.*[16] found that MPs in raw water ranged from 1 to 6614 items/L, and in treated water from 1 to 930 items/L. Fortunately, according to both studies[11,16], drinking water treatment plants (DWTPs) provide an overall removal efficiency of 66.9%-100%, irrespective of treatment types.

Drinking water is a matrix of special concern because it is a source of MP exposure to humans. However, it is not the only route[17]. There are three main routes of entry for MPs to get in contact with the human body: inhalation, ingestion, and through skin[18,19]. Although all three routes contribute to the total amount of MPs to which humans are exposed, ingestion is considered the main source of exposure. Various scientific studies have demonstrated the presence of MPs in the human food chain: in shellfish[20], table salt[21], vegetables and fruits[22], and drinking water[23-27]. However, drinking water has been little studied in comparison to other water bodies.

In parallel, the last decades have witnessed an exponential evolution in the diversity and quality of analytical methods to determine MPs, which has led to significant gains in selectivity and sensitivity as well as decreases in the determinable particle size[3,4,9,10,28-32]. Optical techniques continue to play an important role in the identification and quantification of microplastics, but there is a need to confirm their chemical composition by vibrational or chromatographic techniques. There is still a lack of rationality in the development of robust analytical methods in a systematic way, and their harmonization and standardization need to be addressed. The size of the MPs isolated is dependent on the pore size of the net, sieve, or filter used to isolate the MPs, and this size conditions the amount detected. In the density separation, the density of the selected solution conditions the density of the microplastics that could be separated, and denser MPs sometimes remain with the sample. The sensitivity (in size and amount) of the selected method for the identification of MPs could become another limitation for the comparability of methods. However, several issues depend on the type of water sample in terms of sampling, and identification are better defined now than a few years ago. However, this comparison is constrained by the few methods published thus far.

The objective of this systematic review is to present the severity of microplastic pollution in drinking water with an in-depth analysis of the analytical methods used to establish it. We discuss how the remaining analytical challenges could influence the reliability of the results. To achieve this, a systematic review of studies regarding MPs concentration in different geographical areas was performed. Additionally, we intend to: (1) evaluate the MP occurrence in different drinking water sources (rivers, lakes, tap water, and bottled water) to determine the risk of different exposure origins; (2) summarize the most used analytical methods for MP detection in water samples; and (3) evaluate the possible impacts of MPs on human health.

METHODS

To perform a literature review, the PRISMA guidelines were followed. A search was done for scientific articles in publications databases Scopus (www.scopus.com) and Web of Science (https://apps.webofknowledge.com/). Different combinations of keywords were applied as the criteria of selection for this review in both databases. With the objective of selecting the most relevant publications covering the topic, the terms were retrieved in the article title without any limitation of data since this topic has only been covered recently. The results are summarized in Table 1 and Figure 1.

Following the search of the articles, an analysis was performed in Endnote to determine whether any articles appeared in more than one search. After examining the results, it was found that one paper appeared in both ("Microplastics" AND "Drinking water") and ("Microplastics" AND "Tap water"), two appeared in both ("Microplastics" AND "Drinking water") and ("Nanoplastics" AND "Drinking water"), and one appeared in both ("Nanoplastics" AND "drinking water") and ("Nanoplastics" and "bottled water"). Once screening to remove duplicates was performed, 71 articles remained. This workflow is schematized in Figure 2.

After that, two articles were discarded because they were written in a language different from those spoken by the authors and one because it was published in an inaccessible journal. The remaining 68 articles were studied; however, 11 of them were excluded from the review because their content did not align with this review's objectives. These articles study the efficiency of several wastewater treatments for the removal of MPs and NPs at the laboratory or pilot scale and using spiked samples and/or specially marked MPs. Thus, 57 articles were finally included in the present review.

RESULTS

Of the 57 studies selected, 22 articles cover reviews of the already published literature, one is a

Table 1. Number of references found according to the keywords and to the database used

	Number of articles		
Keywords	**Scopus**	**WOS**	**Total**
"Microplastics" AND "Drinking water"	43	41	46
"Microplastics" AND "Tap water"	7	7	6
"Microplastics" AND "Bottled water"	7	13	12
"Nanoplastics" AND "Drinking water"	7	7	5
"Nanoplastics" AND "Tap Water"	1	1	1
"Nanoplastics" AND "Bottled Water"	2	2	1
			71
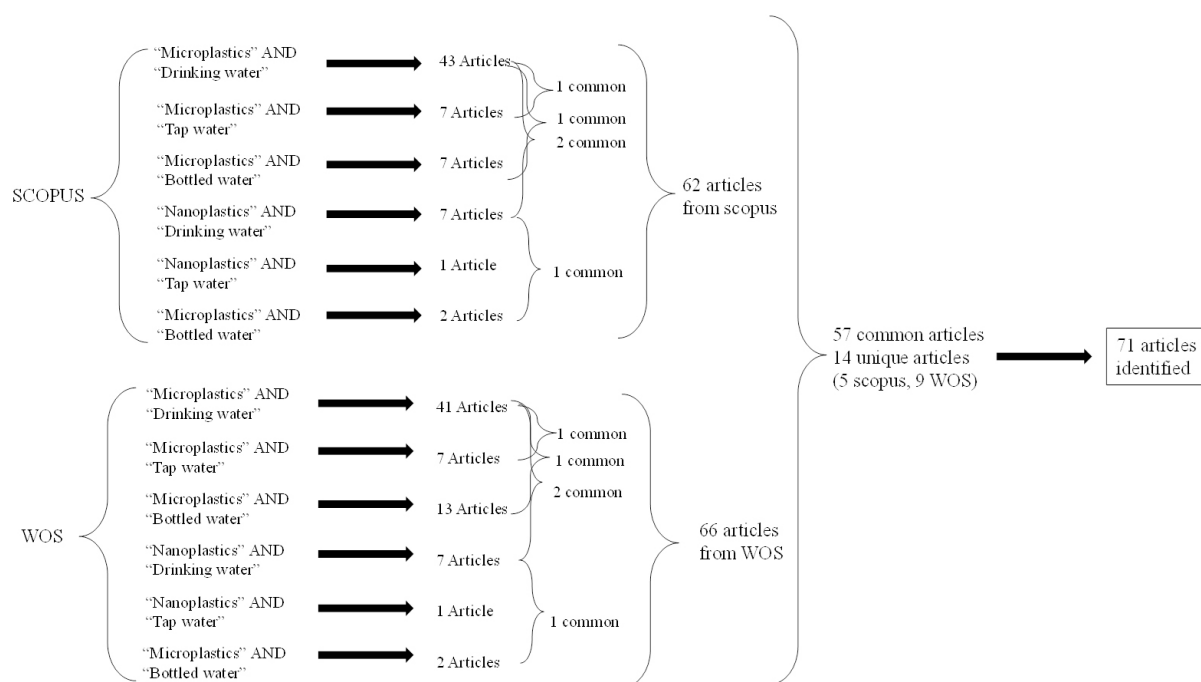
Figure 1. Graphical representation of the search strategy.

corrigendum[33], two pertain to an interchange of comments to an already published manuscript[34,35], and 32 are experimental studies. Table 2 summarizes the topics of the reviews that would be used to reinforce the discussion, and Table 3 shows the most important characteristics of the studies analyzed.

DISCUSSION

Considering the type of review articles compiled in Table 2, there is a concern to highlight what should be known about MPs (or NPs)[5,36]. This knowledge can be divided into five different sections: (1) how to determine MPs; (2) the characteristics of MPs in drinking water, e.g., how many are present and where; (3) how drinking water treatments affect MPs; (4) which facilities release MPs into water; and (5) whether MPs are toxic to human health. This review analyzes all these aspects.

Analytical methods

From the point of view of the sampling and analytical methodology, the comparison between the articles reviewed is not a simple task due to the variety of types of samples, analytical methods, and purposes of the

Table 2. Summary of the most important topics covered in publications reviewing the existing literature

Topic	No of studies	Reference
MPs and NPs: what do the stakeholders need to know?	2	Smith et al.[5]; Xue et al.[36]
Extraction and identification of MPs and/or NPs and quality of the data	4 (1)	Elkhatib et al.[30]; Koelmans et al.[10]; Praveena et al.[31] 2021; Schymanski et al.[32]
Occurrence, fate, and removal of MPs and/or NPs in drinking water treatment plants	12 (3)	Barchiesi et al.[12]; Chen et al.[16]; Koelmans et al.[10]; Li et al.[11]; Novotna et al.[17]; Oladoja et al.[24]; Oßmann et al.[23]; Shen et al.[37]; Eerkes-Medrano et al.[25]; Shrivastav et al.[2]; Sol et al.[27]
Treatment systems and materials that can release MPs and/or NPs to the drinking water	2	Ding et al.[38]; Xu et al.[39]
Risk to human health of microplastics in drinking water	6 (2)	Hogue[40]; Li et al.[11]; Mortensen et al.[41]; Zhang et al.[21]; Eerkes-Medrano et al.[25]; Hohmann-Jedd[42]

References and number in bold indicate those reviews that cover altogether more than one topic of the table and thus are repeated.

studies. The variations are present in all steps of the studies: the sampling process, its transport, the processes performed in the laboratory, and the analysis.

Currently, MPs (and/or NPs) in drinking water have been determined in a wide variety of countries with almost worldwide coverage. Figure 3 shows the countries where MPs (and/or NPs) have been analyzed, including information on the quantity, shape, size, chemical composition, *etc.*

The sampling sites of these studies included: (1) drinking water treatment plants that treat both surface and groundwater (34% of the study); (2) rivers, lakes, wells, and other reservoirs (59% of the studies, considering that many of those that study treatment plants also study raw water); (3) tap water and water from domestic networks (62% of the studies); and (4) bottled water (25% of the studies).

Raw and treated drinking water can be sampled for MPs in different ways. The simple one is to take a conventional grab sample in glass[26,43,59,60,63,64,67], aluminum[44], stainless steel[56], or even high-density polyethylene (HDPE)[58,66] bottles and process them in a laboratory. This is appropriate for volumes between 1 and 25 L. If a higher water volume is needed (> 50 L), grab samples could be immediately passed through a stainless steel wire mesh of different sizes, and then reduced volume samples are taken to the laboratory[45]. Another common system is the use of an immersive electropump connected to one or a full ramp of stainless steel filtration sieves (commonly, from 3.5 mm to 20 μm), and then reduced volume samples are also obtained[47,49,70]. To avoid contamination, the full ramp of stainless steel filtration sieves can be connected by metal junctions to stainless steel valves that collect the water samples from gravity feeds or pressurized supplies[6]. Tap water has also been collected using the water supply cabinets established to perform other conventional analyses in drinking water or within the pipes and lines by in-line filtration of high volumes through nylon or stainless steel filters placed in different holder designs[48,52,55,68,71]. Although nets are the most described way to sample other types of surface and marine waters, the use of plankton nets

Table 3. Summary of the experimental studies analyzed in this manuscript

Area	Quantity (MP/L)	pe/size	Chemical comp.	Sampling	Extraction and analysis	Reference
DWTPs Tehran (Iran)	971 (± 103) 2808 ± 80	< 10 µm (65%-87%)	PP PET PE	Dark glass bottles with a capacity of 2.5 L	Wet Oxidation H_2O_2 Filtration, density separation $ZnCl_2$ (1.55 g cm^{-3}) SEM and µ-Raman	Adib et al.[43]
BW and Tap water in 5 regions (Saudi Arabia)	1.9-4.7	25-500 µm	PE PS PET	24 PET single-use BW 2 glass bottles 2 18 L hard PC plastic container 2 Tap water	Vacuum-assisted filtration with an inorganic filter membrane (0.2-µm pore size) µ-FTIR	Almaiman et al.[44]
Akureyri Urban Area (Iceland)	---	> 1.2 µm 0.7-1.3 µm	PE PVC	20-25 L collected in glass bottles	Filtration (1,2 µm and 0.7 µm glass filters) → 2 fractions Py-GC-MS	Ásmundsdóttir et al.[26]
Freshwater and treated tap water in Bangkok (Thailand)	0.40-2.40	Mostly < 300 µm	PE PET PP	100 L collected using a stainless steel wire mesh of 50 µm Particles rinsed into a glass bottle by SDS in H_2O_2	Density separation NaCl (saturated) and oxidation ($Fe^{2+} + H_2O_2$) µ-Raman spectroscopy	Chanpiwat and Damrongsiri[45]
Surface water in a DWTP in Ontario (Canada)	42 ± 18 in raw water 20 ± 8 in treated water	Fibers 10-45 µm	PE PET-PEST PP PU	10 L samplesTreated water added with isopropyl alcohol and raw water with 10% KOH	1% surfactant Filtration (PC 10 µm filters) Stereomicroscopy µ-Raman	Cherniak et al.[46]
Llobregat River, Barcelona (Spain)	0-3.60	20 µm-0.5 mm	PE PP	10 L samples	Filtration along the sieves of 3.5 mm, 1 mm, 300 µm, 100 µm and 20 µm mesh. Dissolved in water -Density separation adding 25 g of $ZnCl_2$ (δ = 1.29 g/mL) (if needed) Visual, stereo microscope, µ-FTIR depending on the size	Dalmau-Soler et al.[47]
Sant Joan Despí DWTP, Barcelona (Spain)	0.01	0.098-3.288 mm	PP PES PA PTFE Silicone	60 L by filtering "in-line" through a 47 mm nylon filter of 1 µm	Stereomicroscopy, µ-FTIR	Dalmau Soler et al.[48]
Danjiangkou Reservoir (China)		48 µm-5 mm	PP (45%) PS (35%) PE (20%)	20 L 0-20 cm of depth using a Teflon pump is passed through a sieve of (48 µm)	30% H_2O_2 digestion for 12 h - Filtration through a 0.45 µm microfiber filter paper Optical microscopic inspection	Di et al.[49]
DW of Norwegian urban area	6.1-93.1 µg/m^3	≥ 1 µm	PE PA PET	*In situ* modular filtering sampling devices	Subsequent in situ mild enzymatic proteolysis and oxidation (H_2O_2) Py-GC-MS	Gomiero et al.[6]
South-to-North Water Diversion Project, China	516 items/m^3	Fibers of 0.05-1 mm	PET	100 L SW (0-50 cm) filtered using a plankton net (20 µm) at each site. Reduced samples in glass bottles	Samples were filtered onto a 1.2 µm pore size GF/C glass microfiber membrane 30% H_2O_2 was added to digest the organic matter for 72 h	Huang et al.[50]

| Location	Concentration	Size	Polymer	Sampling	Method	Reference
					Stereomicroscope, μ-Raman and μFTIR	
Southeast Nigeria	1.6-42.83 MPs/0.75 L (92% of samples)	Fragments (77.94%) > granules (19.92%) > film (2.14%)	PE PVC PET PDMS	Plastic bottles (750 mL)	Filtered through a cellulose filter paper, stained with Nile red Optical microscopy and SEM-EDS	Ibeto *et al.*[51]
DWTP Sydvatten Skåne (Sweden)	0-0.022 ± 0.019	Mostly < 150 μm, 32% were < 20 μm	PL (87%) PE (9%)	through a 5 μm stainless steel filter.	Incubation in 5% SDS for 24 h at 50 °C and filtration MPs removed from filter by Milli Q water and 50% ethanol. Density separation: $3Na_2WO_4 \cdot 9WO_3 \cdot H_2O$ Supernatant filtered (5 μm stainless steel filter). μFTIR and Py-GC-MS	Kirstein *et al.*[52]
Mineral and sparkling water (Busan, South Korea)	6-58	Mostly < 300 μm	Not specified	Whole bottles as for consumers	Whole bottles (330-500 mL) separately filtered through PCTE membranes (0.4 μm, 25 mm) μfluorescence after PBN fluorophore staining μ-Raman	Lee *et al.*[53]
Tap water Zhejiang University feed by Jiu Xi DWTP (Eastern China)	1.67–2.08 μg/L	Mostly 58-255 nm (MPs and NPs)	POLYOLEFINS PSPVCPA	20-1000 mL and subjected to four filtration steps continuously (0.45 μm and 200, 100, and 20 nm)	Ultrasonication of the filters Use of 0.1 M HCl at 25 ± 2 °C for 30 min Addition of 30% H_2O_2 at 60 °C for 24 h TEM, ATR-FTIR, AFM-IR, and Py-GC/MS	Li *et al.*[54]
Northwestern part of Germany	0-7 MPs m^{-3}	Mostly fibers of 50-150 μm	PL (62%) PVC (14%), PA and epoxy resin (both 9%) PE (6%)	- = Pumped with filtration with 10 μm SS filter, 10 cm below the water surface with pre-rinsing	Add 0.01 M HCl to remove $CaCO_3$/Fe precipitates Filter and rinse with Milli-Q and ethanol 30% Oxidation with H_2O_2 (24 h, 40 °C). If high Fe_2O_3 density separation with $ZnCl_2$ Microscopic inspection and μFTIR	Mintenig *et al.*[55]
12 cities (Japan, US, France, Finland, and Germany)	1.9-225	19.2 μm-4.2 mm	PSSEBSPP	500 mL collected stainless- steel bottle	Vacuum Filtration H_2O_2 treatment μFTIR	Mukotaka *et al.*[56]
Bottled water Bavarian (Austria)	Single-use PET: 2649-2857 Reusable PET: 4889 ± 5432 Glass 6292-10,521	90% of the detected MPs were ≤ 5 μm and about 40% were even < 1.5 μm	PET PE PP Styrene-butadiene-copolymer	21 brands of bottled water obtained in Bavarian food stores	Add EDTA and SDS and homogenization Vacuum filtration (through an aluminum-coated PC membrane filter pore size 0.4 μm) μ-Raman spectroscopy	Oßmann *et al.*[57]
Riobamba city, Ecuador	Only 19% of collected samples had MPs present	5 mm to 1 μm	-		Vacuum filtration through a cellulose filter with a pore size of 2-4 μm The filtered sample was placed in a Petri dish and Rose Bengal pigment was added Stereoscopy	Paredes *et al.*[58]

| Úhlava River (the Czech Republic)	14-1296	Mostly fragments of < 10 μm	Cellulose acetate PET PVC PE PP	2 L of water in borosilicate glass bottles	Sample acidification by adding 1 M sulfuric acid Vacuum filtration SEM, μ-Raman spectroscopy	Pivokonsky *et al.*[59]
WTPs in urban areas of the Czech Republic	1473 ± 34 to 3605 ± 497 (raw water) 338 ± 76 to 628 ± 28 (treated water)	1-10 μm	PET PP PE	2 L of water in borosilicate glass bottles	Two-step filtration through descending mesh size using PTFE membrane filters SEM FTIR spectrometry	Pivokonsky *et al.*[60]
Indira Gandhi Water Treatment Plant, Kolkata, India	2.75-17.88	(50-100 μm) > (< 25 μm) > (25-50 μm).	Fibers: 52%-59% Films/fragments: 41%-48%	Water was sieved through plankton nets (25, 50, and 100 μm). Reduced samples	MPs were density separated using $ZnCl_2$ (1.80 g cm^{-3}). The upper part siphoned on a filter paper (0.7 μm) Washing with deionized water followed by 30% H_2O_2 digestion Optical microscopy, Nile red staining, fluorescence microscopy, and ATR-FTIR	Sarkar *et al.*[61]
Bottled water, Mississippi (USA)	---	---	---	Consumer bottles of drinking water	filtered onto 25 mm φ, 10 μm pore size, PCTE filters Nile red staining and fluorescence microscopy	Scircle and Cizdziel[62]
XiangJiang River and DWTP (Changsha, China)	2173–3998 (freshwater) 338-400 (raw water) 267-404 (tap water)	Mostly fibers and fragments of 1-10 μm	PE PP PS PET	Picked into clean glass bottles with a volume of 10 L	H_2O_2 oxidation Filtration using a vacuum pump (PTFE filters, 0.22 μm φ) Filters were immersed in HCl 0.02 M to dissolve $CaCO_3$ Density separation ($ZnCl_2$) Stereomicroscopy, μ-FTIR, SEM, and Raman	Shen *et al.*[63]
Mexico City Metro system (Public fountains)	5-10 (45% SS) 13-20 (29%) 22-38 (19%) 60-91 (7%)	3-5 mm (3%) 0.5-1 mm (25%) < 0.5 (50%)	PTT Epoxy resin	Water samples (volume 1 L) into pre-cleaned glass bottles up to overflowing.	Samples were filtered through a nitrocellulose filter (0.22 mm) using a vacuum pump Epifluorescence microscope, SEM-EDS, and μ-Raman analysis	Shruti *et al.*[64]
Bottled water (California, USA)	1000-6000 increasing with each cycle	> 4.7 μm	PET	Bottled water	- The bottles' caps were opened and closed 1, 5, 10, and 15 times before analyzing the number of particles generated per open–close cycle NR dye staining Vacuum filtration Trinocular optical microscope	Singh *et al.*[65]
38 samples of tap water from DWTP (China)	440 ± 275	Mostly < 50 μm	PE PP	Sampling using 1 L HDPE bottle to the point of overflowing	1 L: Vacuum filtration with black PC membranes that are further Nile red staining Another 1 L: Addition of HCl and filtration through Al_2O_3 μ-Raman	Tong *et al.*[66]

Yangtze River, China	~1000 to ~6500	Raw water: ~60% 1–5 μm, ~20% 5-10 μm and the others of a bigger size. Sedimentation: ~80% are 1-5 μm; ~15% are 5-10 μm	Fragments > fibers > spheres PET (55.4%-63.1%) PE (15.1%-23.8%) PP (8.4%-18.2%)	Samples were collected from raw water and the effluent of each treatment process in a pre-cleaned glass bottle	Digestion by H_2O_2 (30%) Filtration through a 5 μm PTFE membrane filter Drying of filtrate Qualitative analysis: μ-Raman imaging microscope system Quantitative analysis: SEM	Wang et al.[67]
Rüsselsheim, Germany	Number of MPs was not significantly above the average blank value	-	PE PET PP PS Other particles	Sampling of 0.25-1.3 m^3 with a modified pressure filter house and a stainless steel membrane of 10 μm and 80 mm φ	Acid digestion by 37% HCl at 50 °C for 48 h Vacuum filtration Analysis by μ-Raman	Weber et al.[68]
BW Germany	75-700	≥ 11 μm	PVC PEST PA PE	Raw and deferrized: 1000 and 1453 L using stainless steel filters (50 and 5 μm) Glass bottles	Density separation with $ZnCl_2$, filter and left with citric acid for 24 h, filter again and suspended with ethanol and filtration with an Anodisc Glass bottles: equal plus an additional Anodisc filtration ATR-FTIR (> 0.1 mm) and μFTIT	Weisser et al.[69]
DWTP in Jiaxing, a city of Yangtze River (China)	Max: 2760.14 ± 408.27 (raw water) Max: 379.24 ± 51.25 (treated water)	Mostly 5-20 μm granular MPs	PP VINYON PE PVC VINYLPA	1 L of water for MPs < 20 μm 100-200 L through stain steel sieves for MPs > 20 μm	Filtration Addition of 30% H_2O_2 Use of 0.45 μm PTFE filters (glass vacuum filtration setup) Metalloscope, μ-Raman	Wu et al.[70]
Qingdao, China	0.3-1.6 (tap water) 0.2 to 0.7 (water sources)	10-5000 μm	RAYON PET	4.5 L brown glass bottle and concentrated through a 50 μm PL sieve	Vacuum filtration through a 0.45 μm nitrocellulose membrane Stereoscope and ATR-FTIR spectroscopy	Zhang et al.[71]
BW in Catania (Italy).	100-3000 μg/L	1.28-4.2 μm	Not studied	For each brand, three bottles were collected	Addition of 65% NO_3H and mineralization for 24 h Addition of H_2O and Cl_2CH_2 and centrifugation Evaporated and resuspension with acetonitrile SEM	Zuccarello et al.[72]

AFM-IR: Atomic force microscope–infrared spectroscopy; ATR-FTIR: attenuated total reflection-Fourier transformation infrared spectroscopy; BW: bottled water; DW: drinking water; DWTP: drinking water treatment plant; FTIR: Fourier transformation infrared spectroscopy; GF/C: glass microfiber filter; HDPE: high-density polyethylene; MPs: microplastics; NPs: nanoplastics; NR: Nile red; PA: polyamide; PBN: 1-pyrenebutyric acid/N-hydroxysuccinimidyl ester; PC: polycarbonate; PCTE: polycarbonate track etch; PDMS: polydimethylsiloxane; PE: polyethylene; PET: polyethylene terephthalate; PL: polyester; PP: polypropylene; PS: polystyrene; PTFE: polytetrafluoroethylene; PTT: polytrimethylene terephthalate; PU: polyurethane; PVC: polyvinyl chloride; Py-GC-MS: pyrolysis–gas chromatography-mass spectrometry; SEBS: styrene-ethylene-butylene-styrene; SEM: scanning electron microscopy; SEM-EDS: scanning electron microscopy-energy dispersive X-ray spectroscopy; SS: stainless steel; SW: surface water; UV: ultraviolet.

is reported very few times for drinking water reservoirs[50,61]. The reason is the low amount of organic matter.
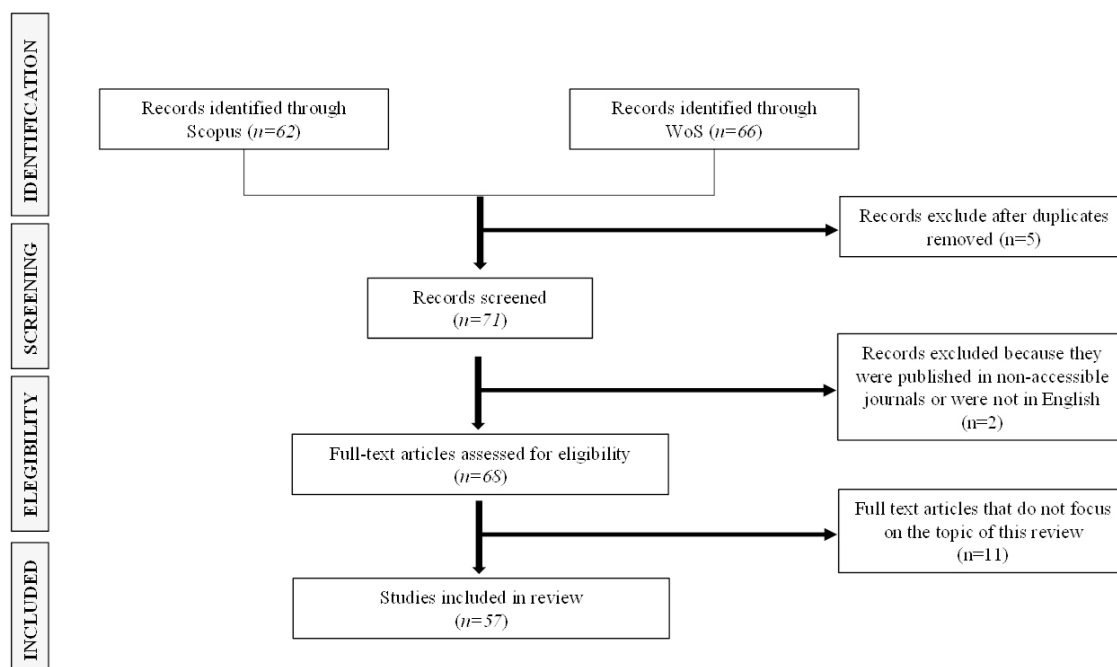
Figure 2. Flow chart of the identification of selected articles.
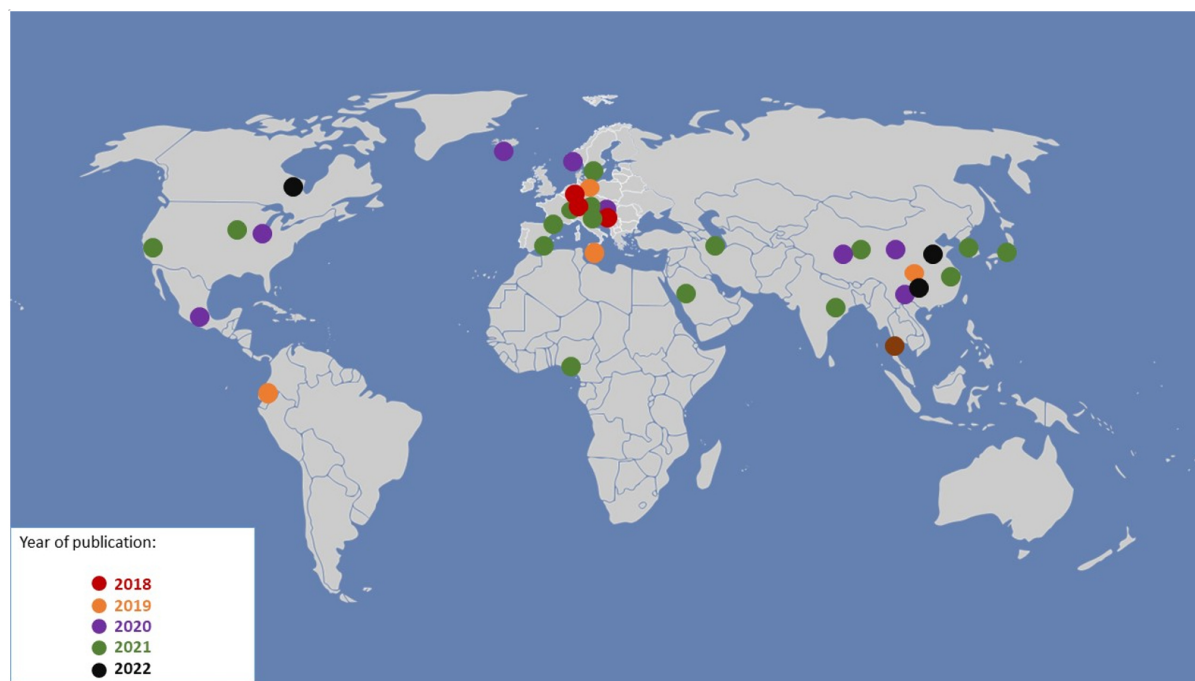
Figure 3. Studies carried out for MPs and NPs in drinking, tap, and bottled water around the world. The colors indicate the year that the study was performed.

The determination of NPs has been reported after the filtration of water by a 0.45 μm conventional filter and then a second filtration through different pore size Anodiscs as small as 20 nm. This sampling procedure attained the simultaneous determination MPs and NPs[69]. However, in comparison with the analytical

methods reported to determine MPs, methods to determine NPs are still scarce, and their analytical performance is still quite unknown.

Grab and reduced samples by passing water through sieves are the most widely accepted techniques for the sampling process. One of the aspects that is recurrently highlighted is the lack of harmonization in terms of the size of the sieves[10,30,32]. The use of different mesh sizes depending on the study complicates comparisons. Another important factor to be considered is the sampling size, which commonly varies between 100 mL and 10 L[31,32] but can reach up to 60 L. The difference found in the content of MPs could also be due to the lack of standard guidelines referring to the sampling size[10]. There is a need for method harmonization to establish the most feasible method. There is a gap in the knowledge about the real need to take reduced samples from large water volumes, and the debate regarding the pore size or mesh size is still unresolved. There is an urgent need to standardize the pore or mesh size to increase the comparability of the studies.

As for the extraction of the samples, the most common methods used are vacuum filtration, digestion, and particle separation by density, as well as the combination of several of them. The pass-through sieves of the sample that arrive in the laboratory have been reported in a few cases[47,60]. Vacuum filtration is mostly the first step of this process. Different pore sizes have been reported, but the most common ones are between 0.2 and 0.45 µm. Water intended for human consumption could be filtrated to a 0.45 µm filter size, since this water has low organic matter content. This ensures the isolation of small MPs. The determination of NPs requires a smaller pore size (< 0.02 µm). This is still hampered by the scarcity of commercially available filters of this dimension as well as the difficulty of coupling filters to the determination methods able to identify NPs. Regarding filters materials, glass microfibers (GF/C)[26,48-50], polycarbonate (PC)[46, 66], polycarbonate track-etched (PCTE)[53,62], aluminum-coated PC[57], cellulose[51,58,64], polytetrafluoroethylene (PTFE)[37,60,67,70], and stainless steel[52] have been described. As a result of the difference between the pore size of the filters used and the flow of the vacuum, findings cannot be analogized. Ethanol and sodium dodecyl sulfate (SDS) are used for both in situ preparation of a reduced sample and laboratory filtration to ensure that all the plastic material is collected in the filter. The filter material is important considering the further identification technique (stained filters, transmission or reflection FTIR, *etc*.). Contamination of the filter by MPs is important, and the adsorption phenomena of MPs in the filters also require further study.

Vacuum filtration is generally followed by density separation, in which samples are mixed with a liquid of a specific density, allowing particles of lower density (MPs) to float, or by further purification using acid digestion, enzymatic digestion, and wet oxidations. Both oxidation and density separation could be indistinctly combined. The most important problem with drinking water is the presence of mineral salts. However, many works do not emphasize this issue[43,44,46-48,55]. Some studies used HCl, HNO_3, or H_2SO_4 to remove $CaCO_3$ and iron or EDTA to sequester mineral salts in general. Strong acids also have an oxidizing effect on organic matter present in water, but the oxidant par excellence is hydrogen peroxide with or without Fe(II) to accelerate the reaction (Fenton reagent). Hydrogen peroxide can be added directly to water[43], although it is most commonly added to the MPs after they have been separated from the sample[45,49,50,54-56,61,70,72]. Furthermore, enzymatic proteolytic digestion has been described once[6]. Density separation involves mostly $ZnCl_2$[37,43,47,55,61,69], NaCl[45], $3Na_2WO_4 \cdot 9WO_2 \cdot H_2O$[52], and the use of organic solvent (dichloromethane)[72]. The recovery of MPs is conditioned by the density of solutions, especially for heavier MPs. There is a lack of knowledge about this recovery depending on the density of the extractant solution.

The next step of the extraction and cleanup is to perform the identification and quantification of the MPs via visualization of the sample through the naked eye, optical microscope, or electron microscope. The visualization of MPs by the naked eye is the least reliable since it has little capacity to detect smaller MPs[47].

Electronic microscopy, especially scanning electron microscopy (SEM), is able to detect the smallest particles[43,59,60,63,67,72], and its combination with energy dispersive X-Ray (EDX) can provide additional information about the chemical composition of the MPs[51,64]. Transmission electron microscopy (TEM) has also been applied, but to a lesser extent[54]. However, the most used technique in the reviewed studies is optical microscopy, especially stereomicroscopy, due to its acceptable capacity for detecting small particles, affordability, availability, and simplicity[46-51,55,56,61,63-65,70,71]. Several studies described the use of fluorescence microscopy to determine MPs, which are not fluorescent, and, therefore, it is necessary to stain them with a fluorophore, such as 1-pyrenebutyric acid N-hydroxysuccinimidyl ester (PBN)[53], Rose Bengal[58], or Nile red[61,62]. However, MPs are many different types of materials, and not all of them are stained with fluorophore with the same intensity. There is also a need to study this aspect in depth.

Polymer chemical identification (and sometimes its quantification) is performed by using μFTIR[44,48,50,52,56,60,63,69], μ-Raman[30,43,45,46,50,53,57,59,64,66-68,70], or Py-GC-MS[6,26,52,54,55]. However, several studies used ATR-FTIR spectroscopy[54,61,69,71] and Raman spectroscopy[63]. In the same way, atomic force microscopy-infrared spectroscopy (AFM-IR), which is able to reveal the functional groups in microzones[54], was reported in one study as advantageous. However, there are not enough studies yet. Another factor that makes it difficult to compare the studies is that the results are expressed in different units of measurement. In some studies, the concentration is given in weight per liter, while in others, it is given as weight in the totality of the sample or the number of particles detected per liter or in the whole sample. However, there is a strong trend to quantify them as MPs/L or MPs/m^3, making the results more comparable.

This review shows that, although there is still a lack of harmonization and standardization, the fact is that most studies use similar sampling and extraction techniques. It is hoped that statistics will soon be established to determine the minimum representative sample size depending on the characteristics of the sampled site and the microplastic content. Likewise, there is an urgent need to carry out interlaboratory tests that will help a lot to have a more harmonized protocol and will provide us with information on the strengths and weaknesses of each determination technique.

Occurrence, fate, and removal of MPs and/or NPs

Bottled water. Seven studies focused on the occurrence of MPs and/or NPs in bottled water. Interestingly, Almaiman *et al.*[44] reported that about 57% of the samples had quantifiable levels of MPs in the size range of 25-500 μm. Ibeto *et al.*[51] found MPs (20-100 μm) in 92% of the samples. Lee *et al.*[53] detected MPs (15-100 μm) in 100% of samples. The amount of MPs found in these studies ranges from 1.9-58 MPs/L. Curiously, Oβman *et al.*[57] had reduced sample size determinable up to 1 μm, and reported two orders of magnitude higher content than previous studies (3659-4889 MPs/L). Zucarello *et al.*[72] decreased the particle size detected to a range of 0.5-10 μm and showed concentrations between 3.16×10^7 and 1.1×10^8 MPs/L, showing the importance of detecting smaller MPs to make studies more comparable. Several types of MPs were detected, but PET (the most common plastic in bottles) and PE (most common plastic in caps) were the main plastic types[44,51,53,57,62,65,70,72]. One of the studies analyzed the effect of water treatment and bottling by determining the concentration of MPs after each step[70] including raw and deferrized water, empty bottles, and caustic cleaning solutions. It was found that each step decreased the quantity of MPs until the final processes of filling the bottle with water and capping, in which a rise in MPs content up to 700 MPs/L was observed. This study showed two interesting points: (1) caustic cleaning solutions contain many MPs, but their carryover to the bottles is prevented by freshwater rinsing; and (2) capping of bottles and their subsequent opening and closing appear to be the main route of entry of MPs. The latter aspect was supported by another study[65]. Oβman *et al.*[57] analyzed the concentration of MPs in different types of bottle packaging. Contrary to what one might think, glass bottles were proven to have the greatest number of MPs/L, followed by reusable PET bottles, and lastly, single-use plastic bottles. The contamination of glass

bottles could be partly explained by the abrasion of the caps on the hard glass bottleneck as well as other processes, such as washing machinery or other steps during the filling process.

Drinking water reservoirs. The reported concentration in drinking water sources is highly variable and depends on the location, the lower size limit of detectable MPs, and the origin of the water (rivers, other surface water, or groundwater). Variations in these characteristics make it difficult to compare one study to another. Some of them described low concentrations already in raw water that range from zero as reported in Iceland[26] where the source is a mix of ground and surface water, up to 42 MPs/L in Canada[46]. However, other studies found higher concentrations, for example 2808 MPs/L in Tehran (Iran)[43], 3065 ± 497 MPs/L quantified in the Uhlava River (the Czech Republic)[59], 3998 MPs/L in the Xiangjiang River (China)[63], and 6500 MPs/L in the Yangtze River (China)[70]. These data demonstrated high variability but global contamination by MPs. It should be noted that China is the world's leading plastics producer, and waste from this industry is not commonly treated. Surface run-off water, water and industrial effluents, and atmospheric deposition can transport microplastics. Thus, factors such as population density and industry dynamics can have a strong influence on MPs concentration.

Drinking water treatment plants. Several articles studied the elimination of MPs in drinking water treatment plants (DWTPs). Conventional DWTPs cannot efficiently eliminate MPs, and some reported results are controversial. Adib *et al.*[43] found that 65%-87% of MPs are smaller than 10 μm. In addition, these MPs were more abundant in treated water than in raw water, which the authors interpreted to mean that conventional DWTPs are unable to remove MPs of this size. The MP removal ability in the investigated DWTPs ranges from 41.2% to 59.0%. Additionally, PP was the most abundant type of MPs in both raw and treated water samples, comprising 27.3% and 24.8%, respectively, and fibers were more abundant than fragments and spheres in raw water (51.1%), while, in treated water, fragments were more abundant than the other two categories (56.7%).

Cherniak *et al.*[46] assessed the elimination of MPs and other particles (> 10 μm) in a DWTP. The treatments were coagulation with aluminum hydroxide, flocculation, anthracite-sand filtration, and chlorination. Samples were also collected from pilot-scale biological filters consisting of anthracite-sand or granular activated carbon (GAC) media operated with or without pre-ozonation and at a range of different empty bed contact times (EBCTs). Full-scale conventional treatment removed 52% of particles. Coagulation, flocculation, and sedimentation presented the highest removal (70%) of any individual unit process. However, the overall removal efficiency decreased to 52%, which is attributed to the effect of airborne particle deposition that occurs while the water remains stagnant (exposed to the atmosphere through ventilation) for disinfection. Most of the particles (> 80%) were identified as 10-45 μm fibers; the MPs were composed of polyester (PL). None of the pilot plant configurations examined improved microplastic removal efficiency compared to conventional full-scale filtration. Interestingly, this study, similar to the previous one, also established that the removal efficiency of conventional treatment may be limited when considering smaller MPs.

These reported results fully agree with those compiled in other review articles about the efficiency of MP removal[2,10-12,17,23-25,27,68]. It is hoped that, in the future, new treatments will improve the efficiency of MP removal and to avoid a decrease in MPs removal efficiency due to post-treatment contamination of the water by aerial deposition and other natural phenomena occurring in the treatment plants.

Dalmau-Soler *et al.*[47] studied MPs in a DWTP and observed that, at the inlet, the mean concentration was 0.96 ± 0.46 MPs/L, with a prevalence of PL and polypropylene (PP), and at the outlet, the mean

concentration was 0.06 ± 0.04 MPs/L, with an overall removal efficiency of 93% ± 5%. Sand filtration was identified as the key treatment for MP removal (78% ± 9%). Furthermore, the results show that ultrafiltration/reverse osmosis (advanced treatment) is more effective for MP elimination than ozonation/carbon filtration.

These observations are coincident with those reported in several reviews on the efficiency of different drinking water treatments, the pros and cons of which are summarized in Table 4. Although these treatments were not specifically designed for MPs and many of them have been found in treated water[12], DWTPs can achieve an overall MP removal rate of 69.9%-100%[16].

Some reviews cover the application of new treatments in water treatment plants, such as electrocoagulation, membranes, or magnetic extraction[2,12,23,25,27,68]. However, these treatments are still in the pilot study stage, and there are no data on their efficiency in eliminating MPs.

Preliminary migration tests performed in several elements of DWTPs indicate that some old and worn elements could be a potential source of MPs, but no evidence of this has been found under normal working conditions. This last aspect also agrees with several reviews on the occurrence of MPs in DWTPs[38,39]. One aspect to consider in the future may be the use of inert materials in DWTPs.

Tap water. Tap water always presents a lower concentration of MPs than the water that enters the DWTPs because the treatments applied to water effectively remove a high percentage of MPs. However, they can still be detected in drinking water. Most concentrations are < 20 MPs/L, as reported for Saudi Arabia[44], Thailand[45], Canada[46], Spain[48]   ,   Norway[6], Iceland[26], Sweden[52], Germany[55], India[61], Mexico[64], a n d Ecuador[58]. However, concentrations were higher in those places where the raw water has a higher concentration of MPs, such as the Czech Republic, China, and Iran, where concentrations up to 328, 440, and 970 MPs/L have been reported[43,49,50,54,59,60,73]. In any case, these concentrations are lower than those of bottled water. There are different explanations for these differences. One is the lower limit of MP size determined. It has been reported that 90% of MPs are < 10 μm. However, many studies only determined bigger MPs. China, as mentioned above, is the top producer of MPs[49,50,59], while the Czech Republic has the fastest growing plastic production in the European Union[60,73]. Furthermore, Iran produces 3,000 tons of plastic waste, mostly in Tehran. This can also be an explanation of the differences[43].

These results show a promising prospect for human health. Concentration differences in drinking water sources suggest that concentrations are higher in areas where there is a high production of plastics. This, however, requires an in-depth study of the characteristics of the area, as there are many factors. Of concern is that water in plastic bottles may increase its concentration of microplastics due to storage and handling. This requires several studies to assess the problem.

Risk to human health of MPs in drinking water

Many studies have demonstrated the presence of MPs not only in aquatic and terrestrial environments but also in food products that humans consume, such as seafood and beverage[11]. The toxic effects of MPs include gastrointestinal irritation, microbiome irregularities, changes in lipid metabolism, and oxidative stress[74]. The entry routes of MPs and NPs in the human body are inhalation, dermal absorption, and intake[41]. Some studies classified the inhaled MPs via aerosol and dust as a high-risk pollutant. Because of their small size, they can be inhaled and deposited in the respiratory system, inducing lung injuries, inflammatory response, and dyspnea in extreme cases. The skin membrane is an effective way to prevent their entry into the body, but MPs can penetrate through open wounds or hair follicles. The effects of MPs

Table 4. Summary of the pros and cons of different drinking water treatments in the elimination of MPs

Process	Involved processes	MP removal efficiency and characteristics	Reference
Chemical treatment	Coagulation and flocculation		Li *et al.*[11]; Cheng *et al.*[16]; Novotna *et al.*[17]; Shen *et al.*[37]
Coagulation–flocculation by chemical coagulants, such as Al and Fe salts	Charge neutralization and adsorption to coagulants plus hydrolyze into electropositive hydroxyl complexes	Poor removal of large MP particles Removal of 40%	
Physical treatment	Clarification, dissolved air flotation, sand filtration, and membrane filtration	Removal of 29%-65%	
Clarification Sedimentation and/or flotation	Removes suspended solids (mineral and organic) and dissolved organic matter. Downward movement that depends on the aggregation of MPs and flocs.	Removal of 40%-54% coagulation/flocculation + clarification. Complete settlement of MPs > 10 μm, 45%-75% of MPs 5 ≠ 10 μm.	
Dissolved air flotation	Dissolving air in water under pressure and then releasing the air at atmospheric pressure.	Overall removal higher = 82%	
Sand filtration Rapid gravity filters (RGV)	Intercept particles. Particles can be strained by the void spaces in the filter.	Remove 29.0%-44.4% Complete removal by triple filtration	
Membrane filtration Porous and diffusional membranes	Porous membranes retain larger particles than the pore size of the membrane by a straining mechanism.	Problems with membrane fouling	
Disinfection	Effective method to kill pathogenic microorganisms	Low removal efficiency = -9.3% to 6.8%	
Chlorination	Inhibiting the activity of bacterial enzymes	No removal = -0.7% to 6.8%	
Ozonation	Attacking cell membranes of microorganisms	Negative removal = -9.3% to -0.3%	
UV treatment	Destroy DNA	Not reported	

through dermal exposure are not very well studied, so more research is needed in this field[11,21].

However, the most important route of human exposure to these contaminants is due to intake[11]. This risk is currently unpredictable; moreover, these MPs are in addition to those potentially consumed from other sources, such as sea salt, beer, food, and seafood. According to the WHO, men should consume 3 L and women should consume 2.2 L of water or water-derived beverages per day[44]. Considering the maximum concentration of MPs in most tap water (< 20 MPs/L) and the recommendation of the WHO, the corresponding daily exposure to MPs would result in 0.7-1 MPs/kg b.w.[6,26, 44-46,48,52,55,58,61,64]. Considering the higher levels found in Iran, China, and the Czech Republic (up to 970 MPs/L), MP consumption would be 30-48.5 MPs/kg b.w.[43,49,50,54,59,60,73]. From these results, we conclude that the level of dietary intake of MP from drinking water is low even in the worst-case scenario, and, according to the current state of knowledge, MP from drinking water do not pose any concern to the consumers.

The European Food Safety Authority (EFSA) stated that particles bigger than 150 μm are not absorbed through the gastrointestinal tract of a mammalian body, but those whose size is under 150 μm can be absorbed either in lymph or in portal veins. Scientists estimate that only 0.3% of microplastic ingested is absorbed. However, only those MPs which are 20 μm or less will be able to penetrate into body organs, and those with a size smaller than 100 nm can cross the blood-brain and blood-placental barriers[52].

Despite the evidence of the distribution and abundance of MPs at present, the implication of these particles on human health is not very well established. The risks associated with MP consumption are not only due to the particles themselves but can also be related to the toxicity of the chemical additives the plastic materials contain. These additives, to enhance the shelf-life and improve the physicochemical properties of plastic

products, can be hazardous to human health as they can be accidentally ingested by humans. However, the lack of knowledge of the most widely used additives in the plastic industry makes it even harder to estimate all the potential effects they can have on human health. Some studies proved that some additives such as polychlorinated biphenyls, bisphenol A (BPA), and phthalates can act as endocrine disruptors, causing disorders related to development and reproduction, such as breast cancer, early maturation, and genital defects.

It is crucial to study more deeply the impact of these particles on human health, especially because of their capacity to adsorb toxicants, including heavy metals, pollutants, and organic macromolecules. Due to this, MPs act as transporters of different toxic substances as well as microorganisms.

CONCLUSIONS

This review highlights the challenges and gaps in the analysis of MPs. In the case of drinking water, sampling is most often done by spot samples of about 1 L volume. However, there are studies that obtain reduced samples of up to 60 L of water. These differences should be standardized. The same applies to the pore sizes of both sieves and filters, which determine the number of microplastic particles found.

Some of the extraction or separation methods, such as density separation, still need a deeper understanding of their analytical characteristics, such as recovery, which may depend on the type of microplastic and the density of the solution used.

The methods of determination are better established, but it is important to perform intercalibrations to assess their comparability. It is also important to consider their sensitivity in terms of particle size and quantity.

It is also remarkable the few methods reported for determining nanoplastics. However, progress is being made in the field. It is expected that in the near future, there will be an explosion of these much-needed methods.

The results on the distribution and transport of microplastics show a certain consistency with the most polluted areas in the world. Moreover, they highlight the effectiveness of drinking water treatments, and the safety of tap water is guaranteed. This is not the case for bottled water, which should be monitored in greater depth.

It has not been fully demonstrated that microplastics are toxic to humans, but neither have they been shown to be harmless, so their presence in water for human consumption needs to be considered.

DECLARATIONS
Authors' contributions
Design: Barceló D, Manzoor I, Picó Y, Soursou V
Literature research: Manzoor I, Picó Y
Data analysis: Barceló D, Manzoor I, Picó Y, Soursou V
Manuscript writing: Manzoor I, Picó Y
Manuscript editing: Barceló D, Manzoor I, Picó Y, Soursou V
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