Emerging Contaminants 11 (2025) 100417

Contents lists available at ScienceDirect

Emerging Contaminants

journal homepage: www.keaipublishing.com/en/journals/emerging-contaminants

Synthetic Microplastics in UK tap and bottled water; Implications for human exposure

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A R T I C L E I N F O

Article history:
Received 16 April 2024
Received in revised form
21 August 2024
Accepted 24 September 2024
Available online 26 September 2024

Keywords:
Microplastics
Tap water
Bottled water
Human exposure

A B S T R A C T

There is increasing concern for public health over inadvertent human exposure to MPs due to potential adverse health effects linked to MPs polymeric composition, toxic chemical additives, and/or harmful microorganisms adsorbing onto their surfaces. While numerous studies have reported MPs occurrence and risk in the freshwater aquatic environment and drinking water sources (e.g., rivers, lakes, and reservoirs), the current state-of-knowledge on MPs pollution in drinking water (i.e., tap water and bottled water) remains limited at a global level. This paper provides the first comprehensive study of the occurrence, concentrations, size distribution, shape, and polymer type of MPs in 177 tap water samples from 13 cities in the United Kingdom, as well as 85 samples of bottled water from 17 popular brands, with various packaging materials, on the UK market.

MPs were detected in all tap water samples (range 6–100 MP/L) and bottled water samples (range 12–62 MP/L). Average MPs concentration in tap water (40 ± 16 MP/L) was statistically indistinguishable from that in bottled water (37 ± 11 MP/L). However, the average MPs particle size in tap water (32.4 μm) exceeded significantly ($p < 0.05$) that in bottled water (26.5 μm), indicating the various purification processes applied to bottled water may help remove larger MPs, but raises concern over the potential adverse health effects from exposure to smaller MPs. The most frequently detected polymer types were: polypropylene (PP), polyethylene (PE), and polyvinyl chloride (PVC) in tap water, and PE, PP and polyethylene terephthalate (PET) in bottled water. A strong correlation was observed ($r = 0.68$, $P = 0.049$) between the plastic cap material (PE) and the predominant polymer type in the bottled water. In terms of morphology, fragments and fibres were the most abundant MPs, together constituting 92 % and 96 % of MPs detected in tap and bottled water samples, respectively.

Using EFSA (European Food Safety Authority) recommended daily water intakes, the corresponding exposures to MPs in different UK age groups were estimated. On a body weight (BW) basis, infants and toddlers were exposed (4 *MP/kg BW/day*) at a higher level than adults (1 *MP/kg BW/day*). This raises concern, given the former's incompletely developed immune/nervous systems rendering them at higher risk of adverse health effects from such exposure.

1. Introduction

Agricultural activities, climate change, droughts, population growth, and increasing industrialisation are reducing and polluting current water resources [1]. Human life and sustainable development depend on the conservation of water and the prevention of pollution [2]. Since plastics became a mainstream material in 1945, plastics have become increasingly popular, reaching 57.2 million tonnes in 2021 (EUROPE, 2022). Plastic pollution worldwide results from excessive plastic use and improper waste management. Plastic debris in the environment gradually break down and eventually convert into microplastics, which are polymer-based particles smaller than 5 mm on their longest dimension [3]. Microplastics are ubiquitous environmental contaminants detected in all environmental matrices, including drinking water [4,5].

Humans are inevitably exposed to microplastics (MPs) daily due to the ubiquitous presence of plastics in all facets of daily life [6]. MPs have been detected in different sources of drinking water

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Peer review under responsibility of KeAi Communications Co., Ltd.

https://doi.org/10.1016/j.emcon.2024.100417

including surface, ground and rainwater [7]. However, fewer studies have measured MPs in actual drinking water e.g., tap water and bottled water, resulting in a dearth of information on human exposure to MPs via this important exposure pathway [8].

Human health concerns associated with MPs have grown because of their recent detection in human biomonitoring studies. Human stool samples were found to contain nine types of microplastics, suggesting internal exposure to microplastics [9]. MPs were detected in urine samples of six volunteers from different Italian cities [10]. Moreover, MPs were also found in in human blood [11] and placenta [12], suggesting that microplastics could reach systemic circulation and cross the placental barrier. According to the World Health Organisation (WHO), MPs may cause adverse health effects to humans through particle dislocation to sensitive organs, oxidative stress, as well as release of toxic chemicals used as plastic additives and/or accumulated in biofilms surrounding MPs particles [13].

Current understanding is that human exposure to MPs can occur through a combination of ingestion (food, water, dust/soil particles), inhalation (indoor and outdoor air), and absorption through the skin (cosmetics, fabrics, atmospheric deposition) [14]. Kosuth et al. [15] suggested drinking water to be a predominant route of microplastic ingestion in humans [15].

To achieve the Sustainable Development Goal of maintaining clean drinking water, MPs, and associated risks for human health from drinking water consumption must be reliably assessed [16]. By 2024, the European Drinking Water Directive will include MPs on 'the watch list' of emerging compounds, to address the growing public concern over MPs and their impacts on human health [17].

Little is known about the occurrence, characteristics, and potential risk of human exposure to MPs via drinking water on the global scale. Few studies investigated the presence of MPs in drinking water sources. A study from the Czech Republic reported average concentrations of MPs in treated wastewater effluents from 3 wastewater treatment plants at 338–628 particles/L [18]. Another study on MPs from different raw water sources in the Netherlands found that groundwater had the lowest microplastics concentrations (<1 particle/m^3), while the highest concentration (460 particles/m^3) was found in riverine water [19].

However, information on MPs in drinking water itself (i.e., tap water and bottled water) remains scarce, and the little information available shows large geographical variability. To illustrate, a study conducted in Germany on bottled water reported water in single-use PET bottles contained 2649 ± 2857 MP/L, while glass bottles contained 6292 ± 10521 MP/L [20], while a study of MPs in bottled water from 9 countries including China, USA and Germany reported MPs in the range of 0–14 MP/L in glass bottles and 7–47 MP/L in plastic bottles [21]. Similarly, the average concentration of MPs in tap water samples from China was reported at 440 ± 275 MP/L [22], while that in tap water samples from the Netherlands was <2 MP/L [19].

Currently, very little is known about the role of packaging material on the contamination of bottled water with MPs, or the impact of drinking bottled water on the overall human exposure to MPs and the potential risk arising from such exposure.

Against this background, the present study aims to investigates the occurrence, polymer types, and particle size distribution of microplastics in UK tap water from several UK cities, and bottled water from different commercial brands. The impact of packaging material on MPs concentrations and polymer types in bottled water is evaluated. Furthermore, we estimate daily exposure to MPs via drinking water in different age groups for the UK population. To our knowledge, this is the first paper to study MPs contamination in tap and bottled water in the UK.

2. Materials and methods

2.1. Sample collection

2.1.1. Tap water

A total of 178 tap water samples were collected from 13 different cities in the United Kingdom (Fig.SI-1). The tap water samples were collected from homes, restaurants, cafes, hospitals, and train stations chosen randomly in each city. 500 mL borosilicate glass bottles with metal screw caps were used to collect tap water samples. Each sample bottle was labelled clearly with the sample code, location (postcode), and the water supplier company for the sampled location (e.g., Severn Trent Water, Yorkshire Water, Thames Water … etc). A list of cities, sample numbers, water suppliers, and drinking water sources are provided in Table.1. Tap water samples were transferred in closed boxes and stored in a cool dark fridge (5 °C) until analysis.

2.1.2. Bottled water

Seventeen of the common bottled water brands, including domestic and foreign brands, were purchased from UK supermarkets in 2022. The investigated brands were chosen based on availability and popularity in the UK market, with the aim of covering different types of packaging materials and water sources. A list of the studied bottled water brands is provided in Table 2. Five samples were acquired from each brand, each sample contained 500 mL of natural, mineral, or purified water. Results are provided as average ± standard deviation of the five identical samples from each brand.

2.1.3. Sample preparation and analysis

In the present study, bottled water and tap water were treated using the same protocol to extract MPs particles. Non-plastic materials were used in all steps. The outside of the closed sample bottles were wiped (Kimtech™ plastic-free wipes, Fisher Scientific®, UK) with ethanol (HPLC grade, Merck™, Dorset, UK) five times before opening the bottles. The confined workspace was cleaned with ethanol every day and was kept covered with clean aluminium foil changed daily. Filtration equipment were cleaned before and after sample filtration with soap solution in Milli-Q water (18 MΩ cm − Milli-Q® EQ (7000) Ultrapure Water Purification System, Merck™, Dorset, UK), followed by ethanol then thoroughly rinsed with Milli-Q water, before drying in a plastic-free oven. All sample preparation steps, were performed in a clean room with separate ventilation and under a clean laminar flow fume hood (Air Science® Technologies Ltd. Merseyside, UK), used only for MPs water analysis.

In the current study, tap and bottled water were treated using the same protocol according to the guidelines recommended in a recent critical review [26] and adopting the method reported by Mukotaka et al. [27]. Briefly, five replicates of each sample were analysed together with one blank containing Milli-Q water (18 MΩ cm) treated as a sample. Sample extraction was performed under the laminar flow fume hood via vacuum filtration (Rocker® 300 vacuum pump, Thames Restek™, High Wycombe, UK). Water samples were filtered through an inorganic silver membrane filter (Sterlitech® 13 mm diameter, 0.45 μm pore size) housed in a 100 % borosilicate glass filter holder kit (Millipore® All-Glass filter holder kit, Merck™, Dorset, UK). Sample bottles and caps were rinsed thrice, with 10 mL Milli-Q water (18 MΩ cm) each, with the rinse water passed through the same filter. The filter was then carefully placed on a glass Petri dish (Fisher Scientific®, UK), containing 200 μL of hydrogen peroxide (30 % w/v, Merck™, Dorset, UK) to digest any natural organic matter on the filter. The digestion step was performed at 60 °C for 24 h.

Table 1
Summary of tap water samples.

Name of the city	Sample numbers/Location	Water Supplier	Source of drinking water
Leeds	3 (Cafés), 1 (Train station) 5 (Restaurants), 1 (Hotel)	Yorkshire Water ltd.	Moorland reservoirs, a quarter from rivers, and a quarter from underground boreholes and spring sources [23].
Sheffield	3 (Cafés), 1 (University) 5 (Restaurants), 1 (shopping Centre)		
Birmingham	3 (Cafés), 46 (Homes) 1 (school), 1 (Hospital) 2 (University), 3 (Restaurants), 2 (shopping Centre)	Severn Trent ltd.	Groundwater and rivers [24].
Coventry	4 (Cafés), 1 (University), 5 (Restaurants)		
Stratford upon Avon	3 (Cafés), 1 (Museum) 1 (Bakery), 1 (Hotel) 4 (Restaurants)		
Wolverhampton	3 (Cafés), 1 (Home), 6 (Restaurants)		
Derby	2 (Cafés), 1 (Museum) 1 (Bakery), 1 (Hotel) 4 (Restaurants), 1 (Market)		
Leicester	3 (Cafés), 7 (Restaurants)		
Nottingham	3 (Cafés), 1 (University), 5 (Restaurants), 1 (Hotel)		
London	2 (Cafés), 3 (Home), 3 (Restaurants), 1 (Hospital), 1 (fountain, Train station)	Thames Water ltd.	Various rivers and Groundwater (Water, 2023a)
Oxford	3 (Cafés), 1 (Museum), 1 (Hotel), 5 (Restaurants)		
Manchester	1 (Cafés), 1 (Student accommodation), 2 (Hotel), 5 (Restaurants), 1 (shopping Centre).	United Utilities ltd.	Reservoirs in the Pennines and the Lake District, from Lake Vyrnwy in Wales and from the river Dee, from boreholes and streams [25].
Southport	2 (Cafés), 1 (Hotel) 6 (Restaurants), 1 (Market)		

Table 2
Summary of bottled water samples.

Brand name	Country of origin	Water type	Packaging material
Evian	France	Natural Spring Water	Plastic bottles (PET) and cap (PE)
Brecon Carreg	Wales	Natural Mineral Water	Plastic bottles (PET) and cap (PE)
M&S Still Water	Scotland	Natural Mineral Water	Plastic bottles (PET) and cap (PE)
AQUAPAX Water	Germany	Pure natural mineral water	Plastic bottles (PE) and cap (PE)
Cano Water	Austria	Natural Spring Water	Metal Cans with Resealable plastic lids (PE)
Princes Gate	Wales	Natural mineral Water	Glass bottles metals cap (inner plastic layer (PE))
One Water	UK	Natural Still Spring Water	Plastic bottles (PET) and cap (PE)
Voss Water	Norway	Purified water	Glass bottles and plastics cap (PP)
Radnor Hills water	Wales	Natural Spring Water	Metal Cans
Radnor Hills water	Wales	Natural Spring Water	Plastic bottles (PET) and cap (PE)
Itsu water	UK	Purified/natural spring water	Refill metal flask with metal cap (inner lining of PE)
Belu Sustainable Water	Wales	Natural Mineral Water	Glass bottles and metals cap (inner plastic lining (PE))
Strathmore Water	Scotland	Natural Spring Water	Glass bottles metals cap (inner plastic lining (PE))
Hildon	England	Natural Mineral Water	Glass bottles metals cap (inner plastic layer (PE))
Fiji Water	Fiji (South Pacific)	Natural Water	Plastic bottles (PET) and cap (PE)
Wilko Water	UK	Natural Spring Water	Plastic bottles (PET) and cap (PE)
Nero water	UK	Purified/natural spring water	Refill metal flask with metal cap (inner lining of PE)

Microplastics analysis was conducted using a Perkin Elmer Spotlight™ 400 Fourier-Transform InfraRed microspectroscopy (μ-FTIR) imaging system with remote-controlled stage, coupled to a Spectrum-3™ FT-IR Spectrometer. The system is equipped with SpectrumIMAGE™ and Spectrum MultiSearch™ software. The whole filter was imaged and mapped in reflectance mode. Spectra produced were compared to those from the Perkin Elmer Microplastics library and/or the independent software tool (siMPle®), using a 70 % match threshold and visual peak diagnostics to ensure 'best fit'. Spectra were acquired in the wavenumber range: 4000–600 cm^{-1} with a resolution of 8 cm^{-1} and accumulation of 16 scans through an aperture size of 20 × 20 μm. If the absorption spectra from 2850 to 3000 cm^{-1}, which derives from C-H vibrational stretching, did not appear, the particle was not an organic compound, i.e., non-plastic, when verified against the library spectrum, and excluded from the MPs count (Harley-Nyang et al., 2022 [50], [27]). The number, shape, and size of the MPs on each filter were determined using microscope images taken by the μ-FTIR microscope in imaging mode, with point mode used for verification of MPs at or near the LOD of 10 μm in size.

2.1.4. Quality assurance and quality control

Conducting studies on microplastics requires avoiding background contamination by MPs in the laboratory. Only pure cotton lab coats and nitrile gloves were worn during the whole experiment process. All sample processing was carried out on the laminar flow bench, which was checked regularly and wiped down with 90 % ethanol. Prior to use, all laboratory equipment/consumables were rinsed thoroughly with Milli-Q water.

Water samples were analysed in baches of 5, to check if sample contamination happened during the sample preparation and filtration, one blank sample (Milli-Q water in glass bottle and metal cap) was analysed alongside each batch. Moreover, one recovery sample (comprising Milli-Q water spiked with a known number of PE MPs in a glass bottle and metal cap) was analysed alongside each 20 samples to ensure good recovery of MPs and no interference/loss during sample preparation steps. Results of the blanks and recovery samples are provided in Tables SI–1. In summary, none of the blank concentrations exceeded 5 % of the average MPs concentrations in the respective batch. Therefore, no blank correction was required. The recoveries of MPs in the recovery samples ranged between (80–112 %) indicating good performance of the analytical method (Tables SI–1).

2.1.5. Statistical analysis

All statistical tests were carried out using Excel (Microsoft 360 package) for Windows and the SPSS 29.0.1.0 program (IBM SPSS Statistics for Windows). Statistical significance threshold was set at 0.05. The microplastic abundance and sizes in the samples was expressed as the mean ± SD of five replicates. Shapiro–Wilk tests were performed to investigate the distribution within the generated datasets. Results revealed the data to be either normal or log-normal distributed. Where a log-normal distribution was identified, further statistical tests were conducted on log-transformed data. Comparison of means between two data sets was conducted using student t-test, while comparison of means among more than two data sets was conducted using analysis of variance (ANOVA) followed by the appropriate post-hoc test.

3. Results and discussion

3.1. Concentration of MPs

One hundred seventy-seven tap water samples and eighty-five bottled water samples were analysed for MPs in this study. Microplastic particles were detected in all tap and bottled water samples.

3.2. Tap water

The concentrations of microplastics in all tap water samples from 13 UK cities ranged from 6 to 100 MP/L with a median of 38 MP/L (Fig. 1a, Tables SI–2)). One-way ANOVA revealed the average MPs in tap water from Birmingham (50 ± 17 MP/L) was significantly higher ($P < 0.05$) than those in sampled tap water from Coventry (32 ± 8 MP/L), Sheffield (29 ± 15 MP/L), and Leicester (30 ± 9 MPs/L). No statistically significant differences in MPs concentrations in tap water were observed among the remaining cities investigated in the present study (Tables SI–2).

To further understand the potential underlying causes of the observed spatial variability in MPs concentrations in tap water from different UK cities, we statistically compared the mean MPs concentrations based on the regional water supplier company (Fig. 1b–Tables SI–3). Statistical analysis (ANOVA) revealed mean concentrations of MPs in tap water samples supplied by Severn Trent Water ltd. (43 MP/L ± 17) were significantly higher than those in water samples supplied by Yorkshire water (32 MP/L ± 15) (Tables SI–3). However, none of the national water suppliers revealed specific measures to remove MPs, while all of them declared strict adherence to all UK national drinking water quality standards. It should also be noted that the number of samples supplied by Severn Trent Water ltd. in the current study (n = 117), is much higher than the samples supplied by the other 3 companies (n = 20 each). This is mainly due to the location of the research team in the UK Midlands, with ease of access to cities within this county supplied mainly by Severn Trent Water ltd. It is also worth mentioning that the second lowest mean MPs concentration (30 MP/L ± 9) was measured in samples from Leicester, supplied by Severn Trent Water ltd. (Tables SI–2). Overall, it is difficult to attribute the observed spatial variability in MPs concentrations in UK tap water samples to a specific factor related to the water supplier or the water treatment processes. Instead, it is more likely a combination of factors extending from the source of water (Table 1), through the treatment processes, and to the final consumer through the piping distribution network [14].

Only a few studies have reported on MPs concentrations in tap water worldwide. An international comparison study on MPs in tap water from 14 countries including: Cuba (n = 1), Ecuador (n = 24), England (n = 3), France (n = 1), Germany (n = 2), India (n = 17), Indonesia (n = 21), Ireland (n = 1), Italy (n = 1), Lebanon(n = 16), Slovakia (n = 8), Switzerland (n = 2), Uganda (n = 26), and the USA (n = 36) reported an overall average concentration of 5 particles/L (range 0–61 particles/L) [15], which is generally lower than the concentrations of MPs measured in our study. Another study on tap water from the Czech Republic revealed much higher MPs concentrations (243–684 Particles/L) [18] than those measured in the present study. A more recent study reported similar MPs concentration (50 ± 17 Particles/L) to those measured in our study, in a tap water sample from Beijing, China [28]. Another study from China reported concentrations of microplastics in 38 tap water samples, with a mean concentration of 440 particles/L [22].

However, it should be noted that direct comparison between the results of the few studies on MPs concentrations in tap water should be conducted with caution. This is due to large variation of the methodologies adopted in these studies including: sample treatment processes (e.g., filter pore size, oxidation of organics), visual-based detection methods (e.g., Nile Red staining, SEM, fluorescence microscopy), chemical-based detection methods (e.g., laser direct infrared spectroscopy, micro FT-IR and micro-Raman Spectroscopy), particle size range of MPs investigated, as well as QA/QC protocols adopted (e.g., blank corrections, recovery detection samples). Such variations can substantially impact the reported MPs concentrations in various studies, and make comparison of their results uncertain [14].

3.3. Bottled water

A total of 17 brands of bottled water with different packaging materials were sourced from the UK market and investigated in the current study. The average MPs concentration in all samples was 37 ± 11 MPs/L, with a minimum of 12 and a maximum of 62 MPs/L (Fig. 2, Tables SI–4). While we investigated different types of bottled water packaging including plastic, glass, metal, and cardboard bottles, as well as plastic and metal caps (Fig. 2), statistical analysis revealed no significant differences ($P > 0.05$) in MPs concentrations among the 17 different brands of bottled water examined. The highest median concentration (46 MPs/L) was measured in water packaged in metal bottles with PE plastic caps, while the lowest median concentration (36 MPs/L) was found in water packaged in cardboard bottles with PE caps. Previous studies have reported similar findings, in terms of lack of direct association between plastic bottle packaging and the number of MPs in the bottled water. This was attributed to various potential sources of MPs to bottled water, including the raw water source, atmospheric deposition during the packing process from conveyor belts and storage tanks, production and processing of plastic caps and liners onto non-plastic (e.g., glass) bottles, as well as the potential fragmentation/flaking of MPs from the packaging material [29–31].

Interestingly, a strong correlation was observed (r = 0.68, P = 0.049) between the plastic cap material (PE) and the

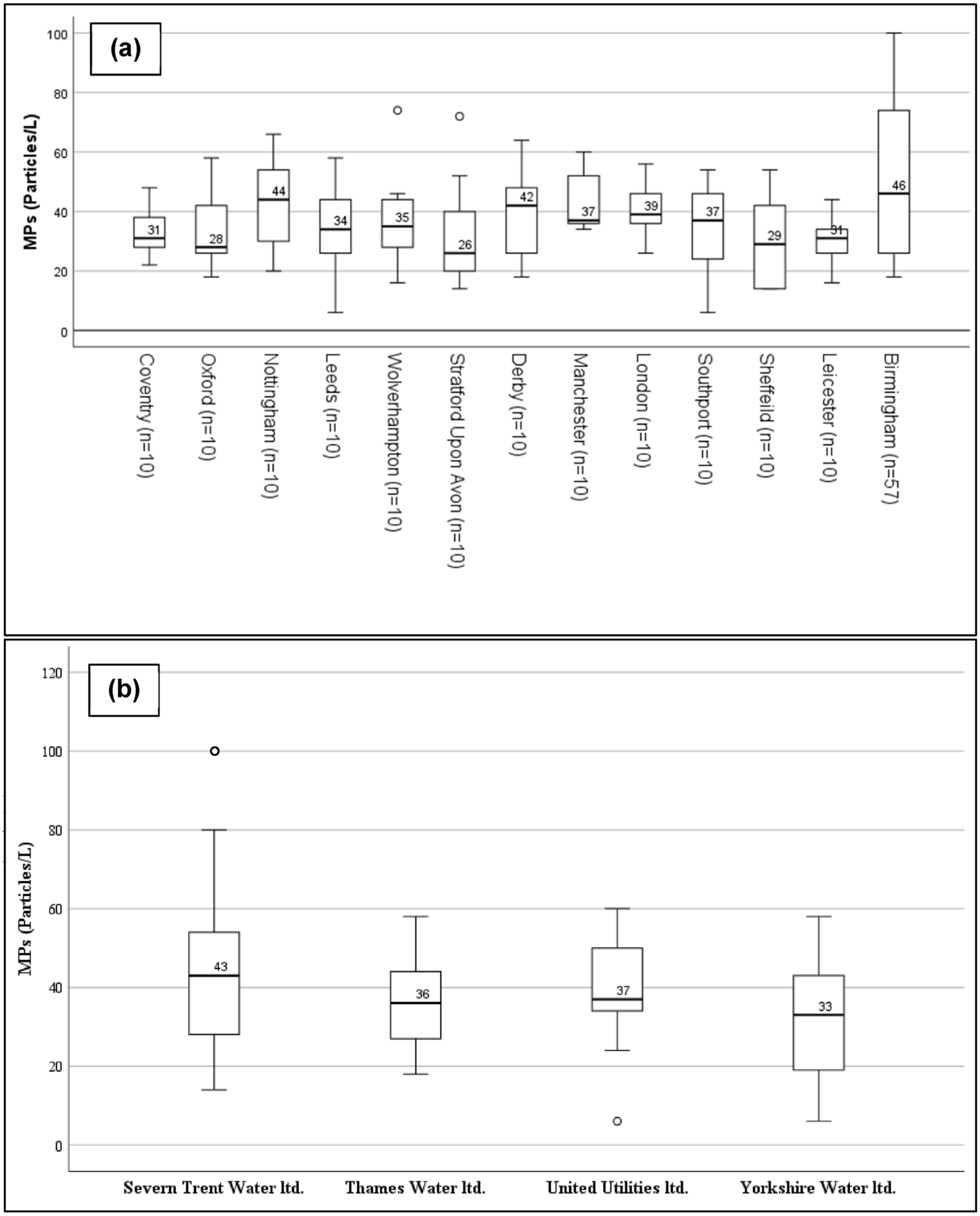

Fig. 1. Concentrations of MPs (Particles/L) in UK tap water samples classified by (a) location from 13 UK cities, and (b) national water supplier (box plots represent 25th and 75th percentile, line and number represent the median, whiskers represent minimum and maximum).

predominant polymer type of microplastics in the bottled water in the present study. Despite the relatively small number of samples in the present study, our finding is in agreement with previous studies which reported that MPs particles may be emitted from caps and seals onto bottled water [8,32].

Notwithstanding the various factors highlighted above, which prevent direct comparison between studies of MPs in water, our results are generally in line with few previous findings from other countries. A study of 34 water samples from Germany reported lower average MPs concentration in 12 samples of single-use PET bottles (2649 ± 2857 MP/L) than in 10 samples of glass bottles (6292 ± 10521 MP/L) [20]. The opposite trend was observed in Thailand where the average MPs concentration in 10 PET bottles (140 ± 19 MP/L) was higher than that in glass bottled water (52 ± 4 MP/L) [33]. Interestingly, a German study of 34 bottled water samples with different packaging materials reported similar

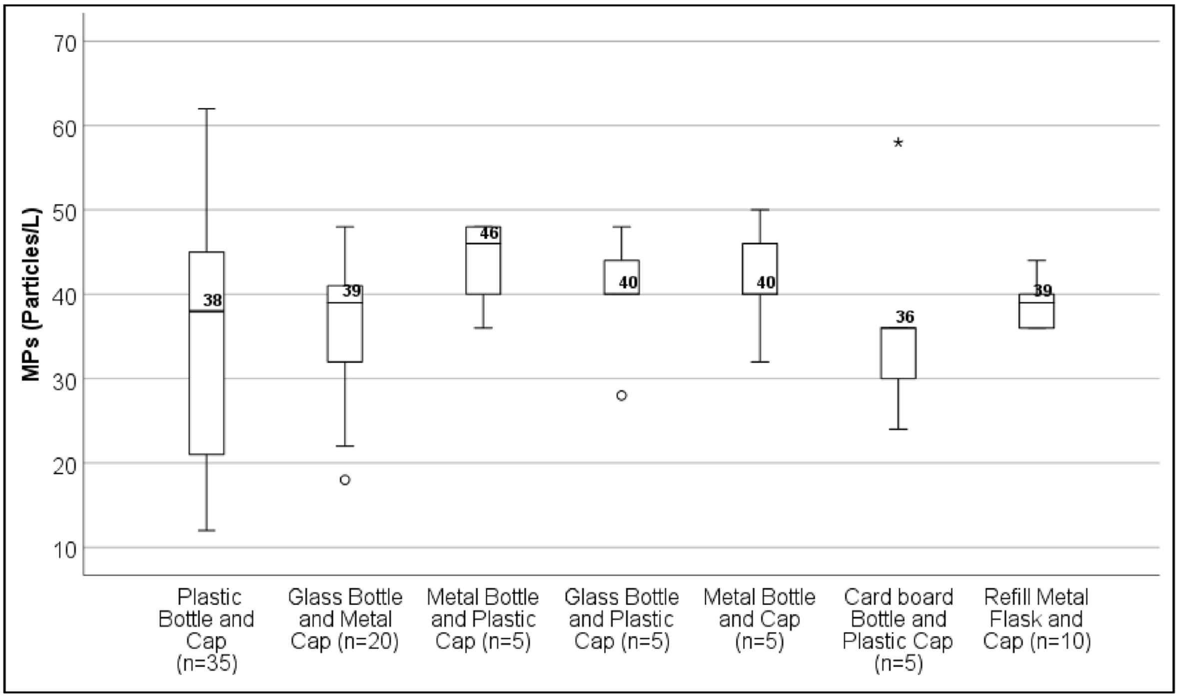

Fig. 2. Concentrations of MPs (particles/L) in 17 brands of bottled water from the UK market, classified by the type of packaging (box plots represent 25th and 75th percentile, line and number represent the median, whiskers represent minimum and maximum).

results to ours, where the lowest average MPs concentrations were found in cardboard bottles with plastic caps (11 ± 8 MP/L), and concentrations of MPs in glass bottles (50 ± 52 MP/L) exceeded those in PET plastic bottles (14 ± 14 MP/L) [34].

3.3.1. Microplastics concentration in tap vs bottled water

In the present study, no statistically significant difference was observed between the concentrations of MPs in the analysed tap water and bottled water samples. The average MPs concentration in all tap water samples (40 MPs/L) exceeded slightly that in all bottled water samples (37 MPs/L), while the median level in bottled water (40 MPs/L) was slightly higher than that in tap water (38 MP/L). A different outcome was reported in a previous study from China, which concluded the microplastics in bottled water (72 ± 45 MPs/L) were higher than in tap water (50 ± 21 MPs/L) [28]. Another study from Saudi Arabia revealed statistically indistinguishable concentrations of MPs in tap water (1 ± 1 MPs/L) and bottled water (2 ± 5 MPs/L) [35]. A recent review article attributed the variance in MPs concentrations among tap and bottled water samples to the different sources of microplastics, including contamination during the packaging process, source of raw water, and relay from the packaging for bottled water, as well as environmental contamination, efficiency of water treatment processes, and relay from the pipes and fittings for tap water [8].

3.4. Characteristics of microplastics in UK drinking water

Microplastics can range in size from microscopic particles in the range of 1 μm to larger particles that are close to the 5 mm threshold [36]. Similarly, previous studies on MPs in drinking water reported different shapes, and few of them reported on the polymer types of the isolated MPs [14,37]. While no clear evidence exists hitherto on the direct impact of different MPs shape and polymer type on their toxic implications to humans upon exposure [37,38], the following sections will briefly discuss the MPs characteristics recorded in the present study for comparability with previous studies. This is of potentially substantial future benefit, when more insight is gained into toxicity of MPs to humans.

3.5. MPs shape

Regarding the shape of microplastics (Fig. 3), irregular-shaped fragments were the most abundant, accounting for 67 % and 72 % of MPs detected in all tap water and bottled water samples respectively. Fibres were the second most abundant morphotype comprising 24 % and 25 % in tap water and bottled water samples, while spheres were only detected at 8 % in tap water and 4 % in bottled water. This agrees with the majority of previous studies on MPs in tap and bottled water, where fragments were the most abundant shape, followed by fibres, then minor contributions from, spheres, films or foams. The abundance of MPs fragments in drinking water was attributed to degradation/decomposition from various plastic products during the processing, storage and/or transfer of drinking water, as well potential degradation of packaging material for bottled water, in addition to atmospheric deposition and other inputs to the raw water source [8,37]. Meanwhile, the regular presence of MPs fibres in drinking water was mainly attributed to the release of synthetic microfibres from domestic washing machines into drinking water sources [18]. Interestingly, a couple of studies have reported different MPs shape profile. In 110 tap water samples from Hong Kong, the majority of microplastics were fibres (98.7 %), with only five films (2.2 %) found in the samples [39], while in 95 bottled water samples from Thailand, MPs fibres accounted for 62.8 % of the total particle content, with the remaining 37.2 % being fragments [33]. There is no clear explanation to the observed variation in MPs morphological profiles from different countries, other than the different MPs input sources to the samples. More importantly, there is as yet no conclusive evidence that the shape of MPs influences their toxic effects [38].

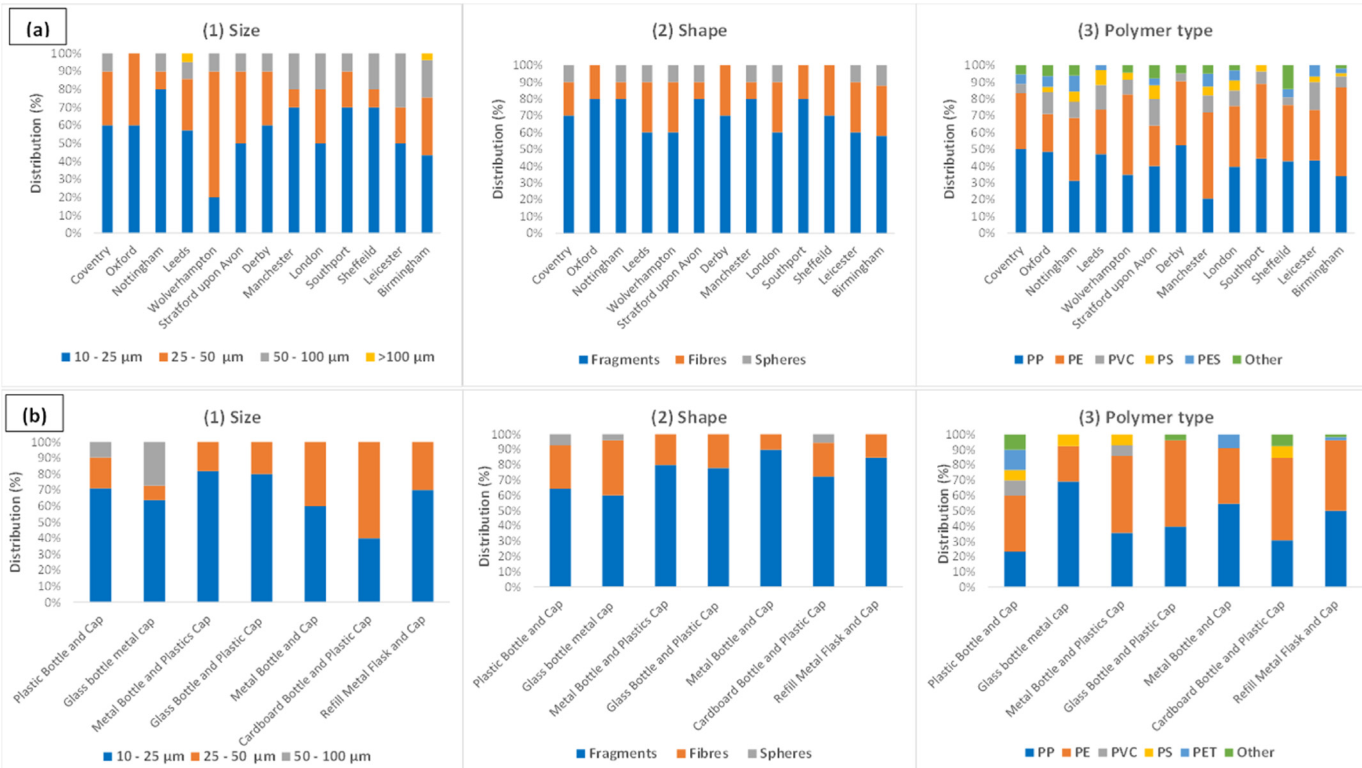

Fig. 3. Distribution (expressed as %) of microplastics (1) size, (2) shape, and (3) polymer type in the studies (A) tap water, and (B) bottled water samples.

3.6. MPs polymer types

Several types of synthetic microplastic polymers were detected in both tap water and bottled water samples analysed in the current study (Fig. 3). Overall, polypropylene (PP) and polyethylene (PE) were the most abundant polymer types in all the studied samples. This is in accordance with previous studies of MPs in drinking water from various countries and is likely explained by the high production volume and extensive application of these two polymer types in consumer products (e.g. PE in plastic bags, bottle caps, and personal care products, and PP in packaging for food, beverages, and cosmetics). Therefore, it is not surprising to detect these polymers in raw sources for drinking water and consequently in tap and bottled water [37]. We detected polyvinyl chloride (PVC) at greater abundance in tap water than bottled water (Fig. 3). This may be explained by the widespread use of PVC in drinking water storage tanks and distribution pipes to domestic settings [8]. Polyethylene terephthalate (PET) was detected at higher abundance in bottled water, particularly in samples packaged in PET plastic bottles (Table 2, Fig. 3). This suggests that least some of the PET fragments detected were derived from the bottle itself. Similarly, PE MPs in these bottled water samples may, at least partially, originate from the PE caps. Polystyrene (PS) was also detected in both tap water and bottled water samples at low percent contributions (Fig. 3). This may be attributed to the abundance of PS MPs in rivers, which are major sources of drinking water [27]. Polyester (PES) fibres were also detected in small numbers in some tap water samples (Fig. 3), which may originate from domestic washing machine discharges to drinking water sources [18].

Other polymer types were detected at very low frequency (<1 %) in the analysed samples. These include polyisoprene, which is not a common polymer in tap water. However, polyisoprene can be used in water filtration systems as a membrane material for removing impurities from water (Vega-Herrera et al., 2023 [51]). Styrene-ethylene-butylene (SEBS), and polymethylmethacrylate (PMMA) were also identified. A survey of existing literature revealed these two polymers were also identified as minor components in a previous multi-national study of MPs in drinking water [27]. The presence of SEBS was attributed to its application in thermoplastic elastomers, asphalt modifiers, and as an alternative to soft PVC, while PMMA was linked to its broad application in medical and dental products.

To date, the toxicological impacts of different MPs polymer types on human health are not fully understood, despite alarming levels of MPs recently detected in various human tissues and excreta [40].

PP: polypropylene; PE: polyethylene; PVC: polyvinyl chloride; PS: polystyrene; PES: polyester; PET: polyethylene terephthalate; other includes: polyisoprene, styrene-ethylene-butylene (SEBS), and polymethylmethacrylate (PMMA), identified at very low frequency.

3.7. MPs size

In tap water samples, MPs ranged from 10.98 µm to 320.39 µm in size with an average of 32.44 µm (Tables SI−5a). Statistical analysis (ANOVA) revealed the average MPs particle size in tap water from Birmingham (49.63 µm) to exceed those from Coventry (22.53 µm), Nottingham (17.03 µm), Leeds (20.35 µm), London (23.77 µm), Sheffield (21.51 µm) and Southport (22.48 µm). It is difficult to find a clear explanation for this observation because the drinking water supplier (Severn Trent Water ltd.) was the same for tap water samples with the largest (Birmingham) and smallest (Nottingham) average MPs particle size in the current study (Tables SI−5a). It should also be noted that the number of tap water samples from Birmingham (n = 57) is higher than that from other cities (n = 10 each), which may have contributed to the observed difference.

In bottled water samples, MPs ranged from 11.36 μm to 98.90 μm in size with an average of 26.49 μm (Tables SI–5b). The largest average MPs particle size (31.47 μm) was measured in glass bottles with metal caps lined with a PE plastic layer (Table 2), while the smallest average particle size (22.00 μm) was found in metal bottles with PE plastic caps (Table 2). No statistically significant differences were observed among the mean MPs particle size in different types of bottled water investigated in the current study (Table SI-b).

Interestingly, the average MPs size in tap water samples (32.44 μm, n = 177) exceeded significantly ($P < 0.05$) that in bottled water samples (26.49 μm, n = 85). More importantly, MPs size distribution among the detected microplastics in all samples was significantly skewed towards the small particle size fraction (Fig. 3). MPs in the size range (10–25 μm) contributed 54 % and 69 % on average to all MPs detected in the analysed tap water and bottled water samples, respectively, while 83 % and 89 % of MPs found in tap water and bottled water samples were below 50 μm in size. This is in agreement with the results of previous studies from Germany, Japan, Hong Kong and China, which reported the concentration of MPs particles in drinking water increased as their size decreased [8,41].

The increased abundance of small sized MPs in the studied samples raises concern over human exposure via drinking water. Current understanding on the toxicity of MPs implies smaller particles display higher toxicity, due to their more facile uptake across epithelial membranes and translocation to various tissues and organs *in vivo,* including the liver, lungs, blood, spleen, and testes [40]. Recent critical reviews on the human toxicity of MPs tentatively identified 20–25 μm as the particle size below which MPs can cross biological membranes, cause oxidative stress and bioaccumulate in tissues and organs, while MPs ≥100 μm were less likely to cause substantial toxicological impacts in human [38,40].

3.8. Human exposure to MPs via drinking water

The estimated daily intake (EDI) is an expression of how many microplastic particles are ingested daily from drinking tap and/or bottled water, according to equation (1) [35,42]:

$$\text{EDI}\ (MPs/kg\ BW/day) = \frac{\text{C}\ (MPs/L)\ x\ \text{DWI}\ (L/day)}{\text{BW}\ (Kg)} \ldots \qquad (1)$$

Where C = Microplastics concentration (MPs particles/L), DWI = Daily water intake (L/day), and BW = Body weight (kg).

To calculate the EDI of MPs via drinking tap and bottled water, we used MPs concentrations measured in the present study, paired with the European Food Safety Authority data on water consumption for different age groups [43], due to the lack of sufficient specific data on the UK population. EDI values were estimated using different exposure scenarios ranging from exposure at the 5th percentile concentration (*best case scenario*) to the 95th percentile concentration (*worst case scenario*) for both tap and bottled water for each age group. To avoid bias due to body weight in the studied age groups, EDIs of MPs via drinking water were estimated on body weight basis (MPs/Kg BW/day) (Table 3), as well as on a daily basis (MPs/day) (Tables SI–6). Few studies have reported on EDIs of MPs from drinking water and even less is known about comparative EDIs among different age groups. Our average EDI for UK adults (1 MP/kg BW/day) is generally in agreement with the average reported for Hong Kong adults (1.2 MP/kg BW/day) [41], while our estimated average EDIs for UK adults and children (Table 3) exceeded those reported for Malaysian adults (0.2 MPs/Kg BW/day) and children (0.25 MP/kg BW/day) from drinking only bottled water [44]. On a daily basis, exposure of UK adults (average 91 MPs/day) and children (81 MPs/day) to MPs via drinking tap water was less than that reported for American adults (630 MPs/day) and children (397 MPs/day) [45]. In a study of particles <10 μm in bottled water samples from Italy, Zuccarello et al. (2019) reported EDIs of MPs for Italian adults and children of 1,531,524 MP/kg BW/day and 3,350,208 MP/kg BW/day, respectively, which are the highest reported to date [46].

Comparing estimated EDI values for human exposure to MPs via drinking water among the few existing studies should be conducted with caution. This is because estimated EDIs may vary considerably depending on a number of factors including the analytical methodology, detection techniques, particle size range analysed, daily water intakes and average national body weights in the different studies [41,44].

Importantly, on a body weight basis, infants and children are more exposed to MPs than adults, despite drinking less water (Table 3). While there is currently no tolerable daily intake or health based limit value for human exposure to MPs, higher exposure in infants and young children raises concern due to the vulnerability of these age groups where the immune and nervous systems are not fully developed, rendering them at higher risk from exposure to xenobiotics than adults [40,47]. Moreover, exposure to MPs incorporates an indirect effect related to the presence of other substances, known as additive chemicals, considered biologically and toxicologically relevant for humans. These chemicals are added to plastic products to achieve desirable properties, which can leach out from the MP particles and become available for human absorption [48]. Several studies reported the presence in MPs of toxic additive chemicals in drinking water, including plasticisers, lubricants, phthalates, flame retardants, and pigments, which may leach out from MPs due to aging, UV exposure, and higher temperatures [8]. MPs can also act as carrier for microorganisms, which may adhere to their surface and be conveyed into the human body [40]. Therefore, the EDIs of the UK population to MPs via drinking water provided in this study raise concerns over the potential toxic implications of such exposure, particularly in infants and young children.

4. Conclusion

This paper provides the first comprehensive study of MPs in UK drinking water, comprising 177 tap water samples from 13 cities, and 85 samples from 17 popular bottled water brands with different packaging. MPs were detected in all tap water samples and bottled water samples. Spatial variability of MPs levels in tap water was observed, with MPs concentrations in tap water from Birmingham significantly higher than those in tap water from Coventry, Sheffield, and Leicester. Mean concentrations of MPs in tap water samples supplied by Severn Trent Water ltd. exceeded significantly those in water samples supplied by Yorkshire Water. The observed spatial variability may be attributed to a combination of factors extending from the source of water, through the treatment processes, and to the final consumer through the piping distribution network. No statistically significant differences were observed between MPs concentrations in the different brands of bottled water investigated, nor among MPs concentrations in the tap and bottled water analysed in the current study. Interestingly, a strong correlation was observed ($r = 0.68$, $P = 0.049$) between the plastic cap material (PE) and the predominant polymer type of microplastics in the bottled water, which indicates fragmentation/flaking of MPs from the plastic cap material as a potentially important source of MPs to bottled water. Also noteworthy is the detection of PET at increased abundance in bottled water samples

Table 3
Estimated Daily intakes of Microplastics via drinking water (MPs/Kg BW/Day) in different UK age groups.

Age group	DWI[a] (L/day)	BW[b] (Kg)	Exposure via tap water (MPs/Kg BW/day)				Exposure via bottled water (MPs/Kg BW/day)			
			Average	Median	5th%ile	95th%ile	Average	Median	5th%ile	95th%ile
Infants aged 6–12 months	0.9	9.2	4	4	2	7	4	4	2	5
Infants aged 1–2 years	1.2	11.4	4	4	2	7	4	4	2	5
Infants aged 2–3 years	1.3	13.8	4	4	2	7	3	4	2	5
Children aged 4–8 years	1.6	22.4	3	3	1	5	3	3	1	4
Girls aged 9–13 years	1.9	43.1	2	2	1	3	2	2	1	2
Boys aged 9–13 years	2.1	42.4	2	2	1	3	2	2	1	3
Adult and adolescent females (≥14 years)	2	67.3	1	1	0.5	2	1	1	0.5	2
Adult and adolescent males (≥14 years)	2.5	80.2	1	1	0.5	2	1	1	0.5	2
Pregnant women	2.3	77.8	1	1	0.5	2	1	1	0.5	2
Lactating (breastfeeding) women	2.7	71.2	2	1	1	3	1	2	1	2

[a] Daily water intake (DWI) data from EFSA [43].
[b] Body weight (BW) data from USEPA Exposure factors handbook [49].

packaged in PET plastic bottles. This suggests PET fragments may have originated from the bottle itself. Fragments and fibres were the most common MPs shapes in UK drinking water, with polypropylene and polyethylene the most abundant polymer types.

The average MPs size in tap water was significantly larger than that in bottled water samples. More importantly, MPs size distribution in all tap and bottled water samples was significantly skewed towards the small particle size fraction (<50 μm). This raises concern because current understanding on MPs toxicity suggests smaller particles display greater toxicity, due to facile uptake across epithelial membranes and translocation to various tissues and organs.

Estimated daily intakes of MPs via drinking water in different UK age groups on a body weight basis revealed infants and toddlers to encounter higher exposure than adults. This is alarming because the incompletely developed immune/nervous systems in infants and toddlers puts them at higher risk of adverse health effects from such exposure. While the toxicological impacts of MPs on humans are not fully understood yet, our study provides important insights into exposure of different UK age groups to MPs via drinking water, which is crucial for accurate risk assessment of this class of emerging contaminants.

CRediT authorship contribution statement

Muneera Al-Mansoori: Writing – original draft, Validation, Methodology, Funding acquisition, Formal analysis, Data curation. **Mia Stephenson:** Validation, Methodology, Formal analysis, Data curation. **Stuart Harrad:** Writing – review & editing, Supervision, Conceptualization. **Mohamed Abou-Elwafa Abdallah:** Writing – review & editing, Writing – original draft, Supervision, Methodology, Data curation, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgement

Muneera Al-Mansoori acknowledges gratefully the provision of her PhD studentship from the *Qatar ministry of higher education*.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.emcon.2024.100417.

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