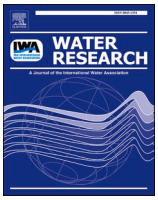
Water Research 255 (2024) 121508

Contents lists available at ScienceDirect

Water Research

journal homepage: www.elsevier.com/locate/watres



Fate and behaviour of Microplastics (> 25μm) within the water distribution network, from water treatment works to service reservoirs and customer taps

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ARTICLE INFO

Keywords:
Microplastics
Tapwater, water distribution network
Water treatment works, service reservoirs
Customer taps

ABSTRACT

Water treatment works have previously shown high efficiency in removing microplastics > 25 μm from raw source water. However, what is less well known is the extent to which microplastics of this size class are generated or lost within the water distribution network, particularly whether there is a greater presence in the customer tap than in the water treatment works outlet. This study focused on the presence of 21 different types of synthetic polymer particles with sizes larger than 25 μm examined through multiple rounds of sampling at outlets of water treatment works (WTW), service reservoirs (SR), and customer taps (CT) managed by seven different water companies in Britain. Nineteen different types of polymers were detected; their signature and concentration varied based on the round of sampling, the location within the water supply network, and the water company responsible for managing the supply. Among the polymers examined, polyamide (PA), polyethene terephthalate (PET), polypropylene (PP), and polystyrene (PS) were the most commonly found. Apart from PET having its highest concentration of 0.0189 microplastic per litre (MP/L) in the SR, the concentrations of the other three most frequent polymers (PS = 0.017 MP/L, PA = 0.0752 MP/L, PP= 0.1513 MP/L) were highest in the CT. The overall prevalence of this size of microplastics in the network is low, but there was a high variability of polymer types and occurrences. These spatial and temporal variations suggested that the MP in the distribution network may exist as a series of pulses. Given the presence and polymer types, the potential for some of the microplastics to originate from materials used in the water network and domestic plumbing systems cannot be ruled out. As found before, the absolute number of microplastics in the water distribution network remained extremely low.

1. Introduction

Plastics possess a unique combination of light weight and strength, exceptional corrosion resistance, and remarkable thermal and electrical insulation characteristics (Andrady and Neal, 2009; Saunders, 2012) making them extremely versatile. The demand for plastic products has shown no signs of slowing down, with a projected increase in global production from 390.7 million metric tons in 2021 to an estimated 590 million metric tons by 2050 (Statista, 2023). This rise in the production of plastic products has challenged the world's ability to manage the resulting waste, and plastic pollution is now a pressing environmental concern of the 21st century (MacLeod et al., 2021; Thompson et al., 2009).

Their relatively low-cost, flexibility and resistance to chemical has allowed them to be selected for some applications in critical infrastructure systems such as drinking water treatment and distribution facilities (Skjevrak et al., 2003; Zhang et al., 2022).

In the absence of a universally agreed scientific definition, microplastics are commonly regarded as plastic particles that are insoluble in water, ranging in size from 1 μm to 5 mm (Frias and Nash, 2019). Primary microplastics are those intentionally produced and added to some products including cosmetics and personal care products (Browne, 2015) for example as microbeads, but most microplastics found in the environment are generated through the weathering or fragmentation of

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https://doi.org/10.1016/j.watres.2024.121508
Received 12 September 2023; Received in revised form 11 March 2024; Accepted 22 March 2024
Available online 23 March 2024

larger plastic products, due to natural physical, chemical, and biological processes (Eerkes-Medrano et al., 2015) and thus termed secondary microplastics. Microplastics in the environment are now considered persistent and ubiquitous contaminants (Junaid et al., 2023; Walker and Fequet, 2023).

The consumption of food and water has been identified as a contributor to the presence of microplastics in the human body (Al Mamun et al., 2023; Kirstein et al., 2021). Whilst there is some evidence suggesting that the accumulation of these particles in mammalian tissues may have negative long-term consequences (Haldar et al., 2023; Kumar et al., 2022; Rubio et al., 2020), the position remains uncertain. There remain major research challenges from the contamination of samples from microplastics in clothing, laboratories and reagents, as well as their close chemical similarity to various natural organic materials. Both issues could lead to over-estimations of microplastics abundance (da Costa et al., 2019).

Combining the detection of particles with the identification of their chemical composition through micro-spectroscopic techniques such as Fourier-transform infrared spectroscopy (μFTIR) and Raman spectroscopy (Raman) remains the most precise method for identifying and quantifying the abundance of microplastics (Schymanski et al., 2021). However, the accuracy of this approach depends on particle size. For instance, in a worldwide interlaboratory comparison involving 22 participating laboratories, those using μFTIR or Raman achieved 92 % recovery and correct identification of pure microplastic standards larger than 50 μm in size (De Frond et al., 2022; Kirstein et al., 2021). This accuracy of numbers of polymer particle detection dropped to just 33 % for particle sizes smaller than 20 μm. Moreover, up to 22 % of μFTIR analyses falsely identified natural materials as plastic, whereas for Raman, this number could reach up to 83 % (De Frond et al., 2022; Kirstein et al., 2021).

Previous research into microplastics larger than 20 μm in drinking water from the outlets of water treatment works revealed a concentration range of 0.00011 to 0.7 microplastic particles per litre (MP/L) (Dalmau-Soler et al., 2021; Johnson et al., 2020; Mintenig et al., 2019). However, in the case of drinking water obtained from customer taps, higher values ranging from 0.02 to 1 MP/L have been reported (Bäuerlein et al., 2022; Dalmau-Soler et al., 2022; Mintenig et al., 2014). This variability in the levels of microplastics between water treatment works and customer taps suggests that there may be additional sources of microplastics within the distribution network.

Previous investigations on the topic focused on water samples collected at a single location and point in time (Bäuerlein et al., 2022; Dalmau-Soler et al., 2022; Mintenig et al., 2014). This limited sample collection makes it difficult to gauge fate and behaviour within distribution networks (Astel et al., 2006; Scanlon et al., 2022).

To make this study as reliable and representative as possible, emphasis was put on a suitable blank correction scheme and a sufficient sampling campaign to assess quantities and frequencies of microplastics larger than 25 μm within water distribution networks. In this case, the focus was on the outlets of water treatment works (WTW), service reservoir (SR) and customer taps (CT). We asked the following questions:

- Are microplastic levels and polymer signatures stable downstream of WTW?
- Are microplastic quantities and signatures different between WTW and SR?
- Is the quantity and microplastic signature at the CT different from other parts of the upstream network?

2. Methodology

2.1. Drinking water network sampled

To achieve a representative coverage of the drinking water supply across Britain, networks from seven water companies were included from England, Scotland, and Wales. Their identities and specific locations were anonymised and are referenced as company A, B, and C etc.

2.2. Collecting water at the outlets of WTW, sr and ct

Our goal was to collect up to 5 m^3 of water within a 24-hour period using a specialized sampling rig. The sampling rig and sampling process involved collecting particles in the drinking water on a stainless-steel filter disc with a pore size of 5 μm and a filter area of 10 cm^2. The filter disc was situated within a 47 mm filter holder made of anodized aluminium and manufactured by Pall Life Sciences Advantec. The filter holder was then connected to the water source using a hose that was approved by the Water Regulations Advisory Scheme (WRAS), specifically a SILEX Platinum Cured Silicone Braided Hose with a 70° Shore hardness and a translucent appearance. The hose, which had an internal diameter of 12.5 mm, was secured using a jubilee clip. Flow meters (2 different models in series to provide redundancy in case of failure) were installed downstream of the filter holder to measure the filtered volume. A full description of the sampling rig is presented in **Fig. S1.** Water was sampled from distribution networks that were managed by each of the water companies on two to three separate occasions between September 2021 and June 2022.

2.3. Controlling microplastic contamination

To minimize contamination during the sampling process, the sampling rigs were only assembled or opened within a safety cabinet equipped with a high-efficiency particulate air (HEPA) filter, which effectively removes 99.999 % of particles larger than 0.3 μm. When equipment or samples were outside the safety cabinet, they were covered with clean aluminium foil. Additionally, to further reduce the risk of contamination from equipment, uncommon plastic substitutes or non-plastic materials were used wherever possible for the sampling and processing equipment. For instance, natural fibre brushes, glass Pasteur pipettes, metal filter rigs, stainless steel, or pure silver filters, fluorinated ethylene propylene/ethylene tetrafluoroethylene (FEP/ETFE) wash bottles, and glass bottles with polytetrafluoroethylene (PTFE)-lined lids and polybutylene terephthalate (PBT) pouring rings were utilised for all sampling and processing vessels.

To minimize contamination from reagents, all reagents utilised in sample processing were filtered through a 0.7 μm glass fibre filter (Whatman GF/F or equivalent) to remove any particulates prior to use. To prevent contamination from synthetic fibres by laboratory operators, all processing activities were conducted by individuals wearing 100 % cotton lab coats. All equipment and glassware were washed exclusively with natural fibre scouring brushes to prevent contamination during cleaning and were rinsed multiple times with 0.7 μm GF/F filtered deionized (DI) water before air drying under foil to avoid airborne contamination. Before sampling commenced, a minimum of 5 L of sample water was run to waste, bypassing the filter, to flush the sampling rig upstream of the filter.

2.4. Preparation of full experimental blanks

Although the arrangement of the standard microplastics field sampling rig with a 5 μm filter was simple, it was necessary to generate experimental blanks that reflected both the field sampling and the laboratory handling. Therefore, we designed a specialised rig to produce what we refer to as "full experimental blanks" . This rig was constructed using six individual stainless steel microplastic filter rigs connected in series, through which we filtered 5000 L of tap water (see **Fig. S2**). The first filter in the sequence collected a microplastic sample as usual and acted as a check of the tap water sample used to generate the experimental blanks. The second stage consisted of two glass fibre filters (1.2 and 0.7 μm pore size) supported by a 5 μm stainless steel filter. This

second stage of the rig was to remove all microplastic particles from the tap water that could otherwise cascade and be captured on the subsequent "blank" filters. The final four filters were stainless steel 5 μm filters, identical to those used in field sampling. Each filter was then processed in the same manner as the field samples. The full experimental blanks were designed not only to reveal any contamination arising from laboratory processing but also any shedding of plastic from within the rigs themselves. We ran this blank process on two separate occasions to generate eight blanks.

In addition to the full experimental blanks, we conducted a series of processing blanks to assess the level of contamination resulting from laboratory cleaning steps. Throughout the project, we ran eight processing blank samples alongside batches of actual samples. These blanks consisted of empty sample beakers processed through the same preparation procedure, and thus only captured microplastics from contamination during processing steps. The level of contamination in these processing blanks was slightly lower than in the full experimental blanks. As a result, we used the full experimental blanks to correct the raw data and calculate the limits of detection.

2.5. Blank correction and limit of detection

For each polymer, the mean blank value was subtracted from the raw count for a sample. This correction was done per volume (m^3) of filtered samples. The limit of detection (LOD) for the blank-corrected sample was defined as 3.3 × the standard deviation (SD) of the blanks as recommended by AOAC International. If the blank corrected value was above the LOD, it was acknowledged as being detected.

2.6. Sample preparation process

After filtration in the field, the rigs were returned to the laboratory and opened within the air filtering cabinet. The 5 μm steel filter with the captured particles was removed with steel tweezers, transferred into a glass beaker, and submerged in 2 % HCl at room temperature for 24 h, after which it was passed through the same 5 μm steel filter and washed with fresh 2 % HCl. This step was necessary to liberate microplastics from mineral precipitates that had formed on the stainless-steel filter disc during filtering ~ 5 m^3 of potable water in the field. Finally, particles were removed from the 5 μm steel filter once more into a 20 mL glass vial using 0.7 μm GF/F filtered DI water. Importantly, we explored a range of weak HCl concentrations and found that 2 % produced the most favourable outcomes in terms of microplastic detection. Subsequently, we evaluated the impact of this 2 % HCl solution on some microplastic standards (PA and PS) and observed no influence on microplastic detection (**Fig. S3**).

2.7. Spike recovery

The effectiveness of recovering microplastics through the sample clean-up processes was evaluated by subjecting DI water that had been filtered through GF/F filters and spiked with a known concentration of polystyrene (PS) and polyamide (PA) standards to the full process. The PA (with size ranges of 30–50 μm and density of 1.13 g cm^{-3}) was produced in-house through cryo-milling and cascade filtration (3000 MP/ml, CoV 11 %), while the PS (with size ranges of 30 – 90 μm and density range of 0.96–1.05 g cm^{-3}) was obtained from Polysciences Europe GmbH, Germany (840 MP/ml, CoV 14 %). We calculated the recoveries as a percentage of the total particles added to the sample. It should be noted that there is currently no established quantitative spike recovery method due to the lack of appropriate certified reference materials for all the different polymers (and sizes).

2.8. Microplastic analysis by μFTIR

For spectroscopic μFTIR analysis, the entire sample was deposited with a glass pipette onto a silver membrane filter (Sterlitech, Washington USA, 25 mm diameter 3 μm pore size) on which a 10 mm internal diameter silicone washer had been placed to define the filtration area. To ensure complete capture of the particles onto the filter, the glass vial was rinsed onto the filter three times after sample deposition. All microplastics within the deposited area of the filter were identified and quantified at a pixel size of 25 μm with the linear array imaging μFTIR spectrometer (PerkinElmer Spotlight 400) set to collect spectra in the range between 4000 and 700 cm^{-1} wave numbers. A background spectrum of the silver filter was collected and removed from the resulting data. Automated spectral matching of the raw data was carried out using the Purency Microplastics Finder software (Hufnagl et al., 2019). This software uses machine learning algorithms to automate the analysis of microplastic measurements, eliminating operator bias that can arise with manual methods (Hufnagl et al., 2019). The output generates particle counts by polymer type and provides information on the two-dimensional characteristics of each particle.

2.9. Polymers quantified

This study reported on the following plastic polymers; acrylonitrile butadiene styrene (ABS); cellulose acetate (CA), ethylene vinyl acetate (EVAc), ethylene vinyl-alcohol (EVOH); polyacrylonitrile (PAN); polyamide (PA); polybutylene terephthalate (PBT); polycarbonate (PC), polyethene (PE); poly-ether-ether-ketone (PEEK); Polyoxymethylene (POM); polyphenylene sulfone (PPSU); Polysulfone (PSU); poly (ethylene terephthalate) (PET); poly(methyl methacrylate) (PMMA); polylactic acid (PLA); polypropylene (PP); polystyrene (PS); polyurethane (PU); polyvinylchloride (PVC); and Silicone (SI). The selection of these polymers was based on their frequent usage, prevalence in the environment, and documented occurrence of some of them in drinking water, as reported by previous studies (Bäuerlein et al., 2022; Dalmau-Soler et al., 2021, 2022; Johnson et al., 2020; Mintenig et al., 2014, 2019).

3. Results

3.1. Blanks and spike recovery

Among the 21 polymers studied, four (PLA, PSU, PPSU, and PEEK) were not detected in the full experimental blanks (**Fig. S4**). Polymers such as PVC, PU, ABS, POM, SI, and PBT were observed on multiple occasions in the experimental blanks. Additionally, some polymers were exclusively found in the experimental blanks and not in the processing blanks. Notably, silicone was commonly observed in the full experimental blanks but absent from the processing blanks. PP and PE, on the other hand, were detected in all of the blank samples. The full experimental blank values were used to calculate the LODs for each polymer.

An assessment of the efficiency of microplastic recovery through the sample preparation and analysis processes revealed a higher recovery rate (91 %) for PS microplastics (90 μm size) as compared to the 32 and 39% recovery rates for the PS and PA microplastics (30–50 μm sizes) standards respectively (**Table S1**). This suggests some numbers provided in this study are likely to be an underestimation.

3.2. Overall microplastic dynamics and abundance

To obtain a comprehensive overview of the distribution and behaviour of microplastics in the water supply networks, both quantitative and qualitative datasets were subjected to multivariate analysis. The quantitative data included concentrations of each of the 19 polymer types detected (above the LOD) across the samples, total microplastic concentrations, and the volume of water sampled. The qualitative data comprised information on polymer types, sampling locations, sampling rounds, and the water companies involved. Principal component analysis (PCA) was performed on 10 components, explaining 99 % of the

variance. Identification and visualization of useful PCA clusters were done using VizRank; a multivariate data visualisation approach guided by Machine Learning and implemented in the Quasar software (Leban et al., 2006; Toplak et al., 2021). The results showed that microplastics distribution could be clustered by whether they came from the 1st, 2nd or 3rd sampling campaign (Fig. 1a), different locations (WTW, SR or CT in Fig. 1b) or by Water Company (Fig. 1c). This indicate that the total concentration of microplastics in drinking water varies based on three factors: (i) the sampling rounds, (ii) the source of the water within the supply network (whether it comes from WTW, SR, or CT), and (iii) the water company responsible for managing the supply (Fig. 1). This result shows how misleading it could be to make generalisation based on one company or one supply round.

Across all sampling rounds and water companies, microplastics concentration ranged from 0 to 0.209 MP/L (with an average of 0.035 MP/L) in the WTW, 0 to 0.165 MP/L (with an average of 0.024 MP/L) in the SR and from 0 to 0.192 MP/L (with an average of 0.027 MP/L) in the CT (Fig. 2a). The frequency at which the highest microplastics concentration within the network was either at the WTW, SR or CTs was assessed. The result shows that the highest level of microplastic occurrence tends to be slightly more associated with the CT (35 %) and SR (34 %) than to the WTW (30 %) (Fig. 2b).

Among the polymers that were examined, PA, PET, PP and PS were the most commonly found polymers in drinking water, occurring in at

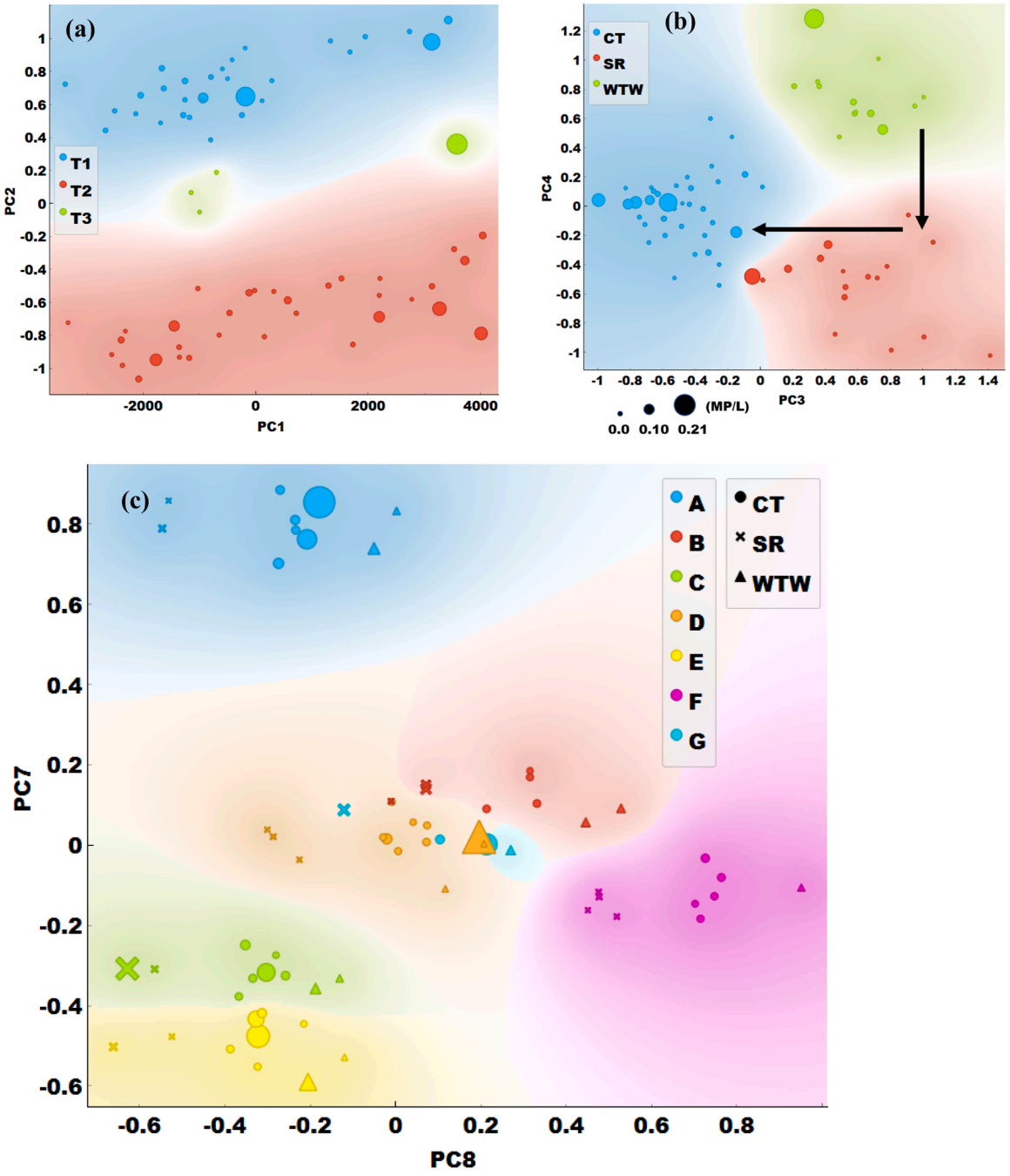

Fig. 1. Prevalence and distribution of total microplastic according to (a) the sampling rounds, (b) the source of the water within the supply network (whether it comes from CT – customer taps, SR – Service reservoir and WTW – Water treatment works), and (c) the water companies (A-G) responsible for managing the supply. T1, T2 and T3 in Fig. 1a are the sampling rounds, the arrows in Fig. 2a indicate the direction of flow and the bigger the size of the symbols in Fig. 1a, b and c the more the microplastics (MP/L).

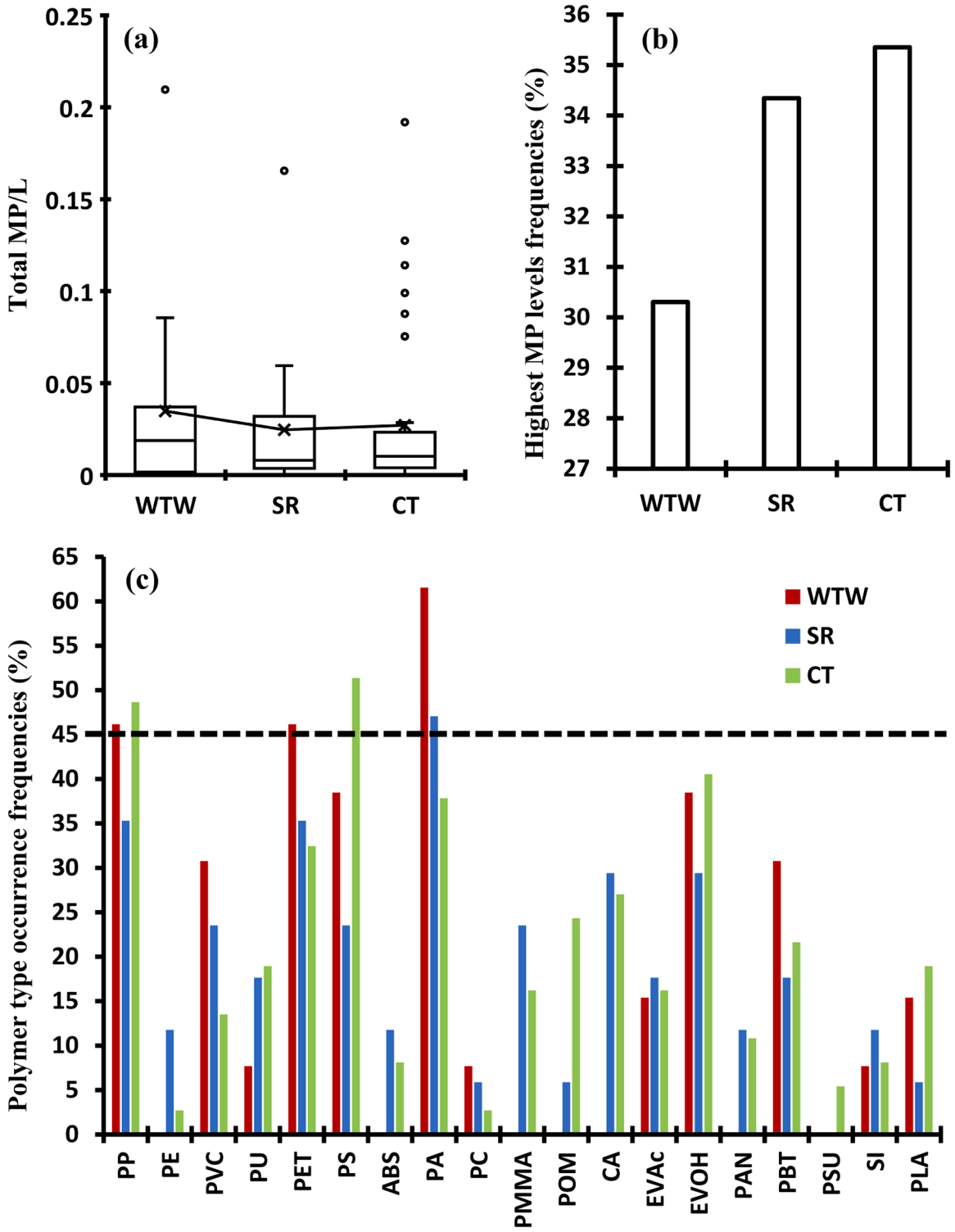

Fig. 2. (a) Total microplastics across all water providers, (b) frequency at which each of the sample types had the highest microplastic level within each network and (c) the contamination frequencies of unique polymer types across the water treatment works (WTW), service reservoir (SR) and customers taps (CT). Boxes show median, Q1 and Q3, and asterisks show means. The figure shows the highest level of MP to be more frequent in CT and PP, PET, PS, PA and EVOH as the most found polymers, occurring in at least 45 % of samples analysed.

least 45 % of samples analysed (Fig. 2c). PA, PET and PP were the most frequently detected polymer in the WTW outlet, while PA was the most prevalent in the SR outlet. PS and PP had the highest frequency in the CT, with their detection occurring in 48 % and 52 % of the samples analysed, respectively. Thus, in terms of polymer signature, the WTW, SR and CT are not identical. To gain a deeper understanding of the nature and behaviour of these most frequent polymers, PCA analysis was conducted on their specific concentrations with respect to the sampling rounds and locations. The result shows both polymer signature and concentrations varied according to the sampling time and source **(Fig. S5).** Apart from PET with the highest concentration of 0.0189 MP/L in the SR, the concentrations of the other three polymers (PS = 0.017

MP/L, PA = 0.0752 MP/L, PP= 0.1513 MP/L) were highest in the CT.

Not all sites were able to be sampled to the ideal quantity of 5 m^3 of water. Consequently, it was crucial to assess whether the results of microplastics prevalence were affected by the varying volumes of water that were sampled. The results of the analysis show no significant association between sample volume and the incidence of microplastics in the samples **(Fig. S6).**

3.3. Distribution of microplastics within individual water supply network

The results of polymer types and abundances according to sampling locations and rounds are presented on a schematic depiction of the network for each of the water companies (Figs. 3-6 and **Fig. S7-S9**). The colour-coded water distribution network in the figures provides information on the specific type of pipe materials found in that company's

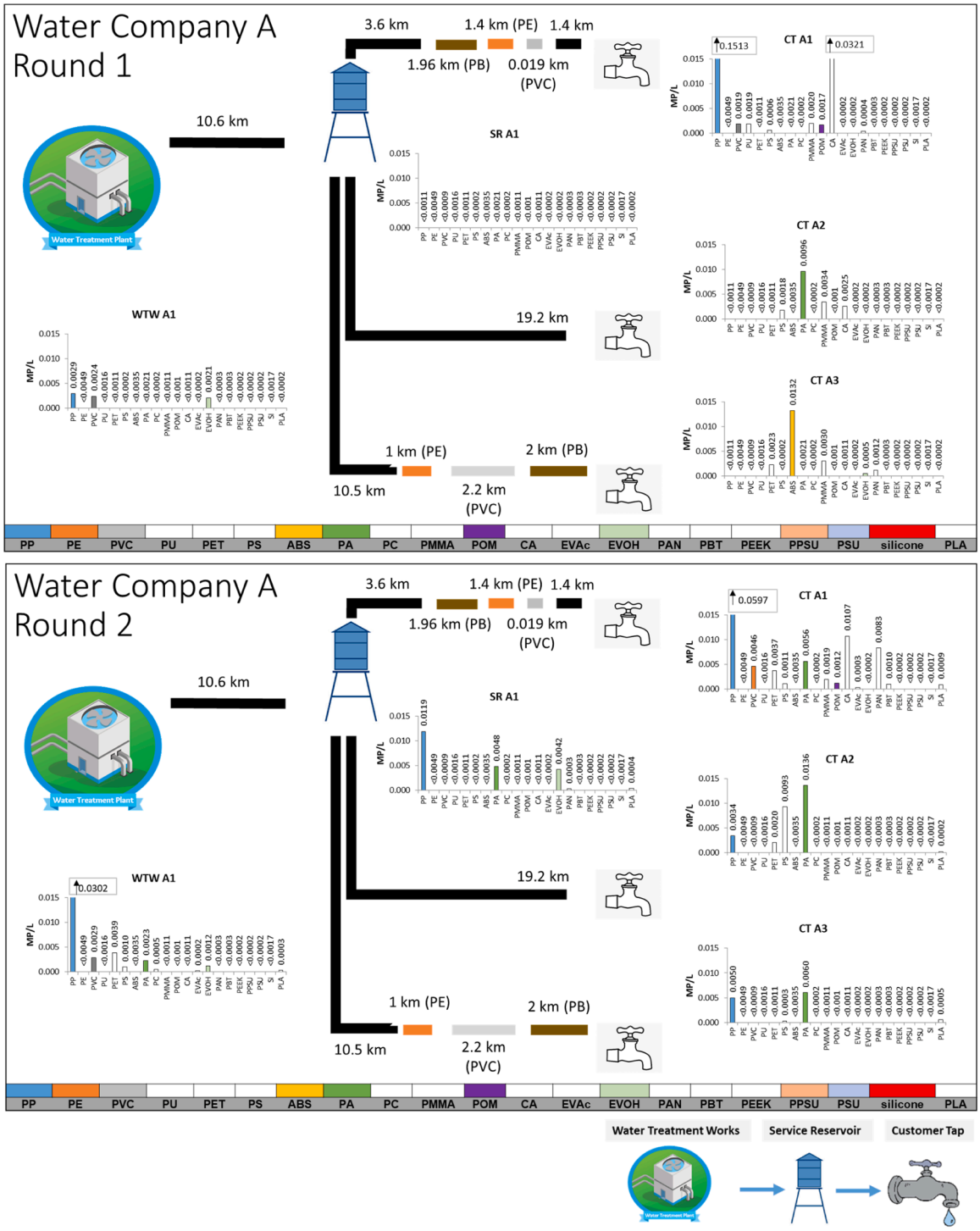

Fig. 3. Prevalence and spatial distribution of microplastics (numbers/L) from the water treatment work (WTW) to the service reservoir (SR) and three customer's taps (CT) in the drinking water supply managed by **Company A** at two sampling rounds. Line connections between sampling locations represent the pipe material between those stages of the network. Black lines are non-polymer pipes. Coloured lines represent plastic piping. Coloured bars represent polymers with plausible sources in the network, while white bars are those without known use in the UK water distribution network. The key at the bottom summarises which colour represents which polymer.

infrastructure. Where data was provided by the water companies, both the distance and material type are presented in the schematic figures. In cases where the pipe is known to be a non-plastic material, black colour is utilised. Grey dotted lines are used to denote areas where details of pipe materials and length were not provided.

In **Water Company** A, the types and abundance of microplastics vary depending on the sampling locations and rounds (Fig. 3). For example, during sampling round 1, PP, PVC, and EVOH were detectable at the WTW outlet, whereas during sampling round 2, additional polymers such as PET, PS, PA, PC, and EVAc were detected in the same WTW outlet. There were no detectable polymers in the SR outlet during sampling round 1, but PP, PA, EVOH, and PLA were detected during sampling round 2. Even though the customer tap CT A3 was situated downstream of pipes made of PE, PVC, and PBT, it did not exhibit any

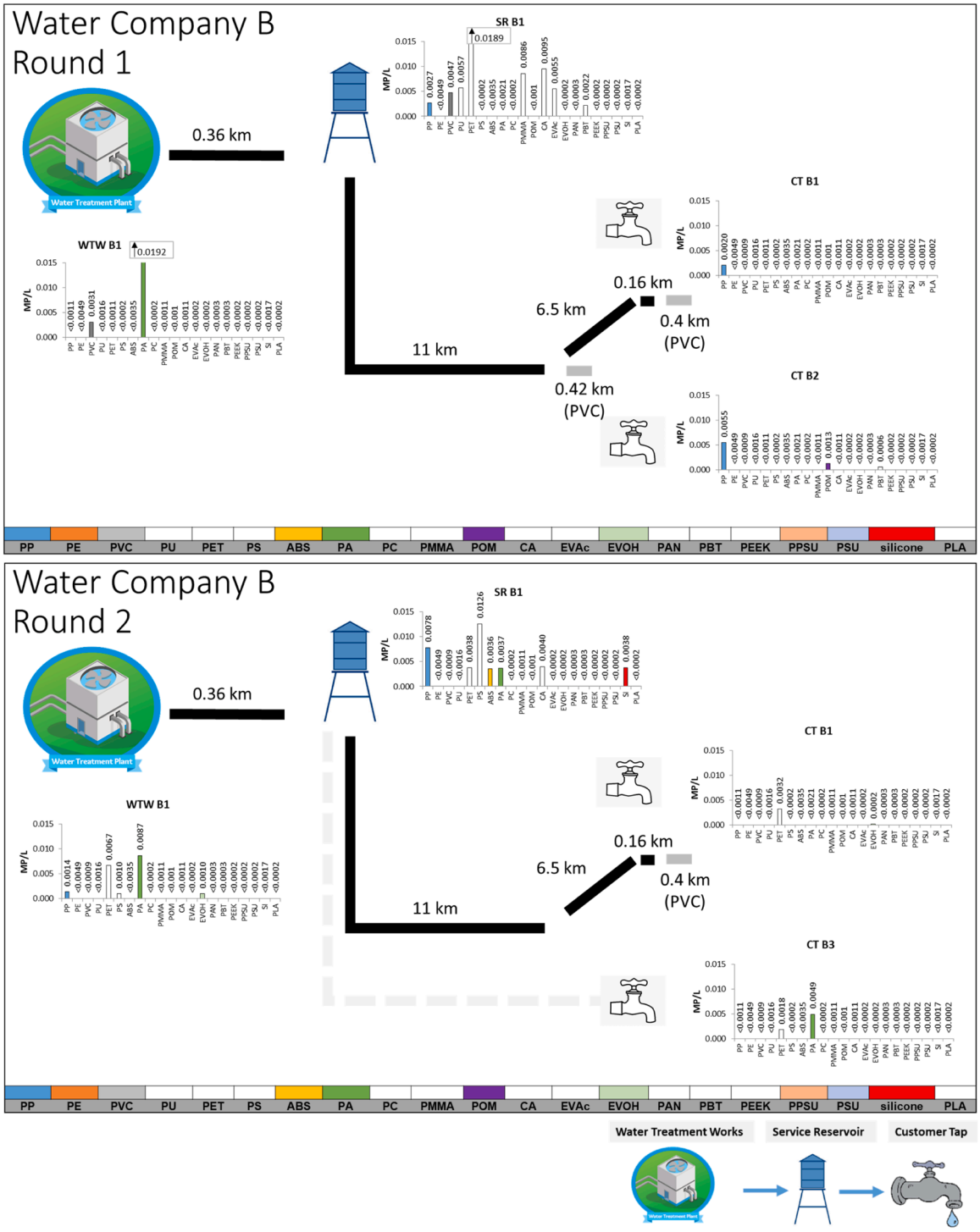

Fig. 4. Prevalence and spatial distribution of microplastics (numbers/L) from the water treatment work (WTW) to the service reservoir (SR) and two customer's taps (CT) in the drinking water supply managed by **Company B** at two sampling rounds. Line connections between sampling locations represent the pipe material between those stages of the network. Black lines are non-polymer pipes. Coloured lines represent plastic piping. Coloured bars represent polymers with plausible sources in the network, while white bars are those without known use in the UK water distribution network. The key at the bottom summarises which colour represents which polymer.

microplastic presence from these polymer types. On the other hand, CT A2, which was downstream of a non-polymer pipe, showed a range of microplastics polymers such as PS, PA, PMMA, and PET. CT A1 consistently exhibited a high level of PP microplastics, with concentrations reaching 0.15 MP/L and 0.06 MP/L during sampling rounds 1 and 2, respectively.

This high PP level was not observed in the other CTs. Overall, Water Company A displayed a consistent microplastic signature from the WTW to SR and CT in terms of the presence of PP, PE, and PA polymers.

Water Company B had a different picture of microplastic presence from the two sampling rounds, with the signature of PP, PE, PET, ABS and PA polymers in the first round in the WTW outlet changing in the second round (Fig. 4). In the second round, the highest microplastics were at the SR outlet but with very little moving down to the CTs.

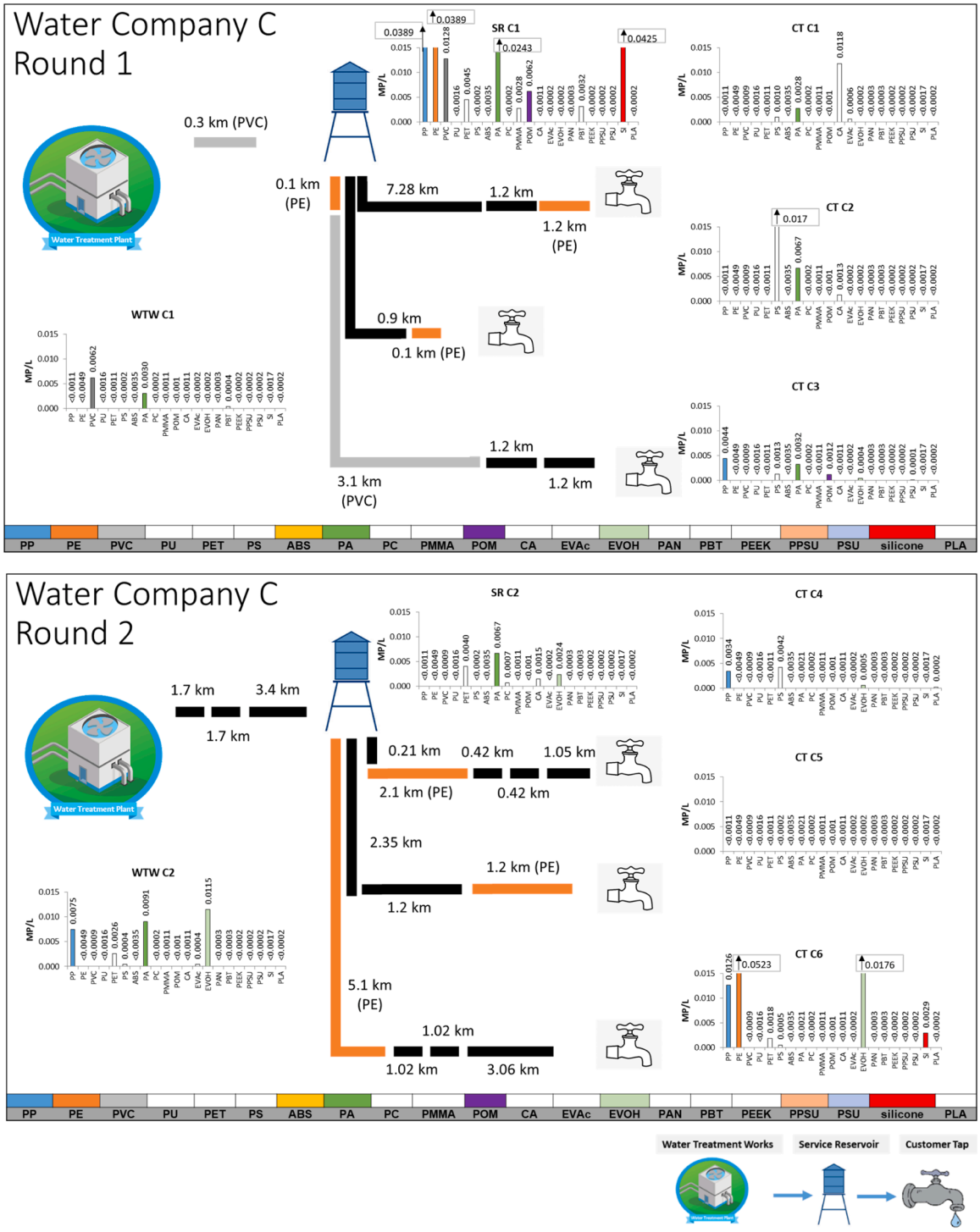

Fig. 5. Prevalence and spatial distribution of microplastics (numbers/L) from the water treatment work (WTW) to the service reservoir (SR) and three customer's taps (CT) in the drinking water supply managed by **Company C** at two sampling rounds. Line connections between sampling locations represent the pipe material between those stages of the network. Black lines are non-polymer pipes. Coloured lines represent plastic piping. Coloured bars represent polymers with plausible sources in the network, while white bars are those without known use in the UK water distribution network. The key at the bottom summarises which colour represents which polymer.

For **Water Company C**, in the first round, SR C1 had the highest level of microplastics (Fig. 5**).** In the second round, the WTW C2 had slightly higher microplastic levels than the first round. Company information revealed that all CTs were fed to some extent by PE for part of the length of the water pipes, but it was only in CT C6 that appreciable PE microplastics could be discovered. This property was supplied by the longest run of PE pipes.

Water **Company D** conducted water sampling from its network on three separate occasions. During the first and second rounds, minimal amounts of microplastics were detected within the network, especially at the WTW and SR (Fig. 6). However, in the third round, elevated levels of microplastics were observed at the WTW D1, specifically SI followed by PA. Despite these higher levels at the WTW, the SR and CTs showed low levels of microplastics. Among the customer taps, CT D2 stood out with a distinctive and consistent presence of polystyrene (PS) microplastics across all three sampling rounds. This presence of PS was not observed in the other CTs, making CT D2 unique in its microplastic composition.

In **Water Company E**, much higher microplastic was present in sampling round 2 compared to 1 for the WTW, whilst the opposite was true for the SR **(Fig. S7)**. The CTs were relatively low in microplastics apart from CT E3 which was the highest on both occasions. Although slightly different sites were examined in the two sampling rounds at **Water Company F**, the presence of microplastics was relatively very low across the network **(Fig. S8)**. Microplastics in the network managed by **Water Company G** was only examined in one sampling round. Unlike some of the other sites, something of a network polymer signature could be traced from WTW to SR to CTs centring on PP, PE and PA. The CT G3 tap had the greatest presence of microplastics **(Fig. S9).**

4. Discussion

Despite significant efforts to reduce microplastic contamination during water sampling and processing, the analysis of the blanks reveals that background contamination still occurs and needs to be taken into account. For example, the sampling piping was made of SILEX platinum-cured silicone braided hose, which was chosen as a relatively uncommon and durable hose material approved by water companies and regulations. Nevertheless, the occurrence of silicone exclusively in the full experimental blanks, in contrast to the laboratory processing blanks, implies that the sampling rig and the filtering of large volumes of water through silicone tubing generated unique microplastic contamination. The presence of PP and PE in all the blank samples suggested a common source of contamination during the sampling and processing stages of the procedure. As these polymers were not present in any of the equipment used, it is possible that they were introduced from an ambient source within the field and laboratory environment. These results highlight how vital it is, particularly when trying to detect microplastic in clean matrices like potable water, that blanks replicating the full sampling and processing steps are designed (Johnson et al., 2020; Koelmans et al., 2019).

Providing sufficient blank values allows a robust LOD to be devised (Bertil and Örnemark, 2014). In this study, we opted for a LOD calculation that theoretically yields a significance level of 0.05 for both false negative and false positive error rates. This implies that if a sample contains a concentration of microplastics at the same level as the LOD, the likelihood of a false positive result, where the field sample actually contains no microplastic, is only 5 % (Armbruster and Pry, 2008). While blanks can help prevent the occurrence of false positives, spike recovery is important in identifying false negatives and providing information on the number of microplastics that might have been overlooked. In this study, the recovery rate for PS and PA standards with sizes ranging from 30 to 90 μm was between 32 and 91 %. Despite its potential benefits, the utilisation of positive control (spike recovery) in drinking water microplastics analysis remains uncommon (Koelmans et al., 2019). In the few studies where spike recovery was documented, diverse polymer types and sizes were employed. In a study utilising PA particle (63–90 μm diameter), the recovery rates for raw and portable water microplastics extraction protocols were 101 ± 27 % and 113 ± 15 %, respectively, closely aligning with the recovery rate reported for PA in this study (Johnson et al., 2020). Another study investigating microplastics in drinking water utilised PE (average diameter 100 μm) and recorded a recovery rate between 65 % and 80 %. (Bäuerlein et al., 2022). Unfortunately, the lack of appropriate standards for different polymer types (and subtypes – e.g., different colours, different amounts, or types of additives etc.) in varying size ranges limits the effectiveness of spike recovery. Thus, positive controls can only be considered indicative. In addition to the meticulous blank correction and positive control implemented in this study, our research adhered entirely to the quality standards prescribed for the analysis of microplastics in freshwater and drinking water (Koelmans et al., 2019).

The average concentration of microplastics > 25 μm reported in this study is comparable to those documented in previous studies of WTW and CT in Germany, Spain, and the Netherlands (Bäuerlein et al., 2022; Dalmau-Soler et al., 2021, 2022; Johnson et al., 2020; Mintenig et al., 2014, 2019). However, in this study, we show that microplastic particles are not uniformly dispersed in drinking water in time and space.

It was difficult to see within the one to three sampling rounds, a consistent pattern in the prevalence and distribution of various plastic polymers. Certain plastic polymers appeared dominant in one sampling round, only to diminish in the next. Notably, the plastic polymers detected at the WTW did not necessarily align in type and quantity with those found at the SR or CT. Although it was common to encounter microplastics of polymer types that could plausibly originate from different parts of the infrastructure, there was no clear relationship between microplastics abundance and the distances the water travels in plastic pipes from the WTW to the CT **(Fig S10).** It seems difficult to escape the conclusion that microplastics of many different polymers are ubiquitous even in the cleanest of environments.

It was surprising to find microplastics of different polymers at the SR that were not evident at the upstream WTW and similarly between SR and CT. Other than concluding that the integrity of the network from WTW to CT was breached at certain points, it may be that occasional episodes, or surges, of microplastics move through the system as a pulse from the WTW. Several plausible scenarios may be applicable:

1. The distribution network remains secure from external contamination and does not itself leach microplastics. In this case, the only source of microplastics would be from WTW-treated water, perhaps as occasional events (pulses).
2. The distribution network remains secure from external contamination and does not itself leach microplastics. However, microplastics originating from the WTW become attached to the pipe surfaces and occasionally slough off, leading to pulses of microplastics discharge.
3. The distribution network is secure but itself leaches microplastics from pipework and junctions, including within the customer's house.
4. The distribution network is compromised by the intrusion of microplastics between WTW and customer tap, perhaps at the SR, where new microplastics enter the system.

To rule out any of these possibilities, further investigation would be required, entailing a more comprehensive sampling approach, a rigorous assessment of pipework integrity, and an examination of the nature of local plumbing works or repairs, where relevant, at the CT.

The health requirements for water intake vary among individuals based on factors such as age, level of physical activity, and climate. In the United Kingdom, the National Health Service recommends consuming 6–8 glasses of fluids daily, which equates to approximately 1.2 L (NHS, 2021). However, according to the European Food Safety Authority (EFSA), females aged 14 and above should drink 2.0 L per day, while males aged 14 and above should drink 2.5 L per day (EFSA Panel on Dietetic Products and Allergies, 2010). In the context of this study,

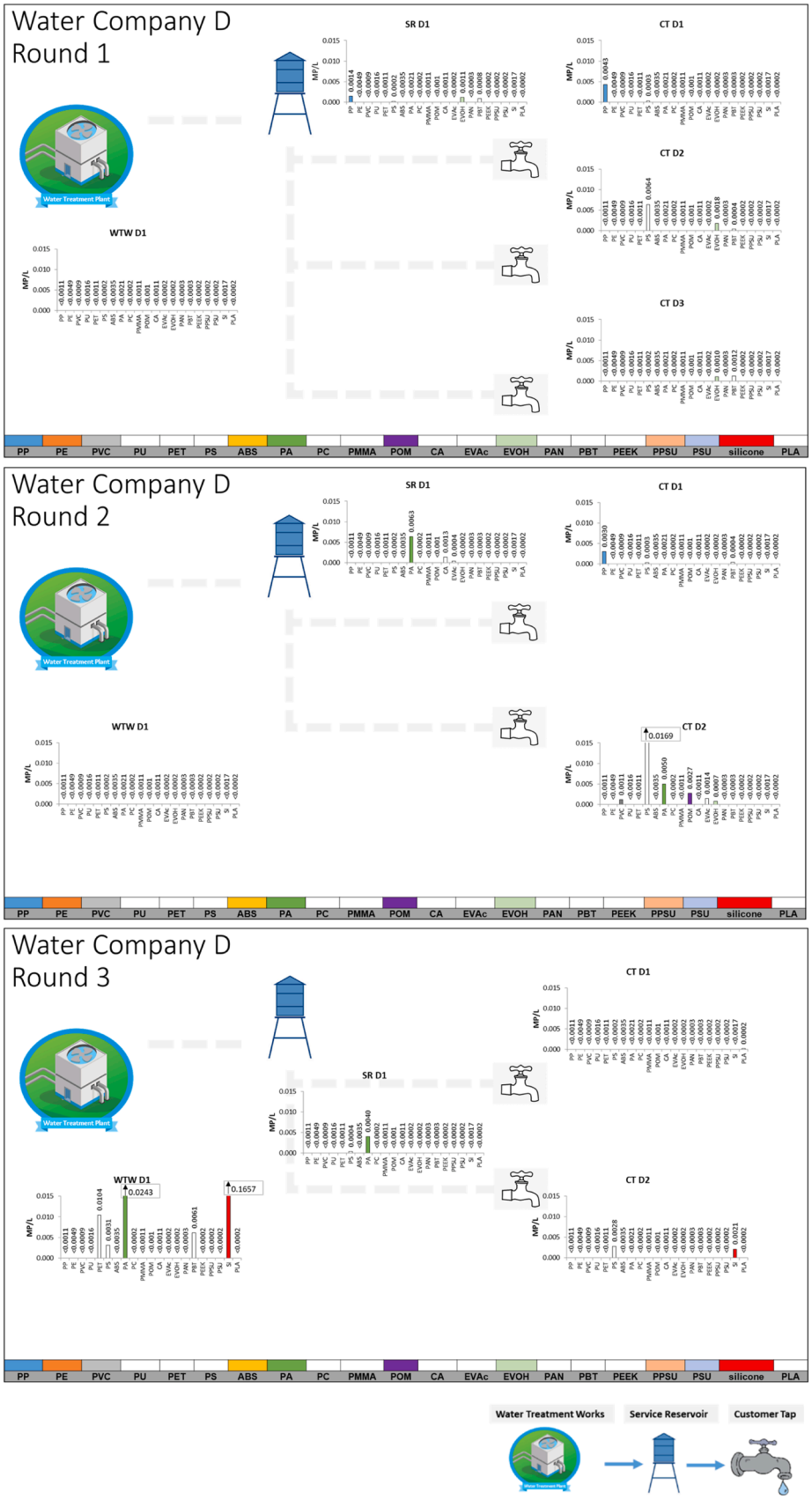

Fig. 6. Prevalence and spatial distribution of microplastics (numbers/L) from the water treatment work (WTW) to the service reservoir (SR) and three customer's taps (CT) in the drinking water supply managed by **Company D** at three sampling rounds. The pipe material used for connecting the sampling points is unknown. Coloured bars represent polymers with plausible sources in the network, while white bars are those without known use in the UK water distribution network. The key at the bottom summarises which colour represents which polymer.

considering the average concentration of this class of microplastics at 0.0276 MP/L and an assumed daily water intake of 1.2–2.5 L, it would require approximately 14 to 30 days for an adult consuming tap water at the recommended rate to encounter 1 microplastic (> 25 μm) particle. However, in scenarios where the maximum concentration of microplastics (0.2096 MP/L) is considered, encountering 1 microplastic (> 25 μm) particle could happen within a shorter span of 2 to 4 days of drinking tap water. At present, the understanding of the significance of various polymer types, particle sizes, quantities, or concentrations with respect to the hazardous properties of microplastics is limited. Hence, it is premature to make any remarks regarding the potential risks associated with microplastics in drinking water.

To give some context on exposure, the average concentration of microplastics in bottled water has been recorded at levels spanning from 8.5 ± 10.2 to 4226 ± 3385 MP/L (Kankanige and Babel, 2020; Makhdoumi et al., 2021; Ossmann et al., 2018; Samandra et al., 2022; Schymanski et al., 2018), with reports of tea having an average of 11 ± 5.26 MP/L, soft drinks having 40 ± 24.53 MP/L, energy drinks having 14 ± 5.79 MP/L, and beer with 152 ± 50.97 MP/L (Shruti et al., 2020). In contrast, this study reports an average concentration of 0.025 MP/L in tapwater. It would appear that human exposure to microplastics larger than 25 μm would be trivial from tap water compared to other commercial beverages. However, it is important to note that the comparison is made with caution, given variations in the sizes and types of microplastics examined, and the analytical methodology employed across the studies.

4.1. Conclusion, limitation, and future works

We employed a comprehensive blank correction protocol, multiple water sampling campaigns and μFTIR analysis to attempt to unravel the types, abundance, distributions, and occurrence of microplastics >25 μm within water distribution networks.

The results revealed that microplastics levels and polymer signatures varied across different sampling rounds and locations downstream of the WTW. Although the highest levels of microplastic occurrence were more frequently associated with the CT and SR than the WTW, the average microplastics levels among these locations were similar. The variability suggested that microplastics might be moving through the system as a series of pulses.

Among the polymers examined, PA, PET, PP, and PS were the most commonly found, with PA, PP, and PS showing their highest concentrations in the CT. Despite our efforts, confident identification of the precise source of these microplastics larger than 25 μm within the network remained elusive. The potential for some of the particular microplastic polymers to originate from materials used in the water network and domestic plumbing systems cannot be ruled out. Nevertheless, it is essential to note that their presence in the tap water network is relatively low, occurring at an average concentration of 0.025 MP/L.

Considering the variability observed in this study, to provide deeper insight into the sources of microplastics within the supply network, future research would need to adopt a relatively high sampling frequency, ideally conducting monthly sampling over a period of one to two years.

The reliable quantification of sub-micron and nanosized plastic particles in potable and tap water is still required.

CRediT authorship contribution statement

Gbotemi A. Adediran: Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Validation, Writing – original draft, Writing – review & editing. **Ruairidh Cox:** Formal analysis, Investigation, Methodology. **Monika D. Jürgens:** Conceptualization, Formal analysis, Funding acquisition, Investigation, Methodology, Writing – original draft. **Elise Morel:** Formal analysis, Investigation, Methodology, Writing – original draft. **Richard Cross:** Formal analysis, Funding acquisition, Project administration, Data curation. **Heather Carter:** Formal analysis, Investigation, Methodology. **M. Glória Pereira:** Conceptualization, Funding acquisition, Methodology, Supervision. **Daniel S. Read:** Conceptualization, Funding acquisition, Project administration, Supervision. **Andrew C. Johnson:** Conceptualization, Funding acquisition, Investigation, Project administration, Resources, Supervision, Writing – original draft, Writing – review & editing.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Andrew Johnson reports financial support was provided by UK Water Industry Research (UKWIR).

Data availability

All data used are presented in the manuscript and supplementary information.

Acknowledgements

This work was funded by UK Water Industry Research (UKWIR), grant number EQ/01/A/248. We would like to thank Project Manager, Nina Jones, the Programme Lead, Christine Murray and members of the steering group for their valuable contributions and guidance on this project. We would also like to thank the staff of the Water Companies for their cooperation during fieldwork planning and for assisting with sample collection.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.watres.2024.121508.

References

Al Mamun, A., Prasetya, T.A.E., Dewi, I.R., Ahmad, M, 2023. Microplastics in human food chains: food becoming a threat to health safety. Sci. Total Environ. 858, 159834.

Andrady, A.L., Neal, M.A., 2009. Applications and societal benefits of plastics. Philos. Trans. R. Soc. B: Biol. Sci. 364 (1526), 1977–1984.

Armbruster, D.A., Pry, T., 2008. Limit of blank, limit of detection and limit of quantitation. Clinic. Biochemist Rev. 29 (Suppl 1), S49–S52.

Astel, A., Biziuk, M., Przyjazny, A., Namieśnik, J., 2006. Chemometrics in monitoring spatial and temporal variations in drinking water quality. Water Res. 40 (8), 1706–1716.

Bertil, M., Örnemark, U., 2014. The Fitness For Purpose of Analytical methods: A laboratory Guide to Method Validation and Related topics. A laboratory Guide to Method Validation and Related Topics. LGC, Teddington, Middlesex, UK.

Browne, M.A., 2015. Sources and pathways of microplastics to habitats. Marine Anthropogenic Litter 229–244. https://doi.org/10.1007/978-3-319-16510-3. ISBN 978-3-319-16509-7.

Bäuerlein, P.S., Hofman-Caris, R., Pieke, E.N., Ter Laak, T.L., 2022. Fate of microplastics in the drinking water production. Water Res. 221, 118790.

da Costa, J.P., Reis, V., Paço, A., Costa, M., Duarte, A.C., Rocha-Santos, T., 2019. Micro (nano) plastics–analytical challenges towards risk evaluation. TrAC Trends Anal. Chem. 111, 173–184.

Dalmau-Soler, J., Ballesteros-Cano, R., Boleda, M.R., Paraira, M., Ferrer, N., Lacorte, S., 2021. Microplastics from headwaters to tap water: occurrence and removal in a drinking water treatment plant in Barcelona Metropolitan area (Catalonia, NE Spain). Environ. Sci. Pollut. Res. 28 (42), 59462–59472.

Dalmau-Soler, J., Ballesteros-Cano, R., Ferrer, N., Boleda, M.R., Lacorte, S., 2022. Microplastics throughout a tap water supply network. Water Environ. J. 36 (2), 292–298.

De Frond, H., Thornton Hampton, L., Kotar, S., Gesulga, K., Matuch, C., Lao, W., Weisberg, S.B., Wong, C.S., Rochman, C.M., 2022. Monitoring microplastics in drinking water: an interlaboratory study to inform effective methods for quantifying and characterizing microplastics. ChemosphereChemosphere 298, 134282.

Eerkes-Medrano, D., Thompson, R.C., Aldridge, D.C., 2015. Microplastics in freshwater systems: a review of the emerging threats, identification of knowledge gaps and prioritisation of research needs. Water Res. 75, 63–82.

EFSA Panel on Dietetic Products, N. and Allergies, 2010. Scientific opinion on dietary reference values for water. EFSA J. 8 (3), 1459.
Frias, J.P.G.L., Nash, R., 2019. Microplastics: finding a consensus on the definition. Mar. Pollut. Bull. 138, 145–147.
Haldar, S., Muralidaran, Y., Míguez, D., Mulla, S.I., Mishra, P., 2023. Eco-toxicity of nano-plastics and its implication on human metabolism: current and future perspective. Sci. Total Environ. 861, 160571.
Hufnagl, B., Steiner, D., Renner, E., Löder, M.G., Laforsch, C., Lohninger, H., 2019. A methodology for the fast identification and monitoring of microplastics in environmental samples using random decision forest classifiers. Anal. Methods 11 (17), 2277–2285.
Johnson, A.C., Ball, H., Cross, R., Horton, A.A., Jürgens, M.D., Read, D.S., Vollertsen, J., Svendsen, C., 2020. Identification and quantification of microplastics in potable water and their sources within water treatment works in England and Wales. Environ. Sci. Technol. 54 (19), 12326–12334.
Junaid, M., Liu, S., Chen, G., Liao, H., Wang, J., 2023. Transgenerational impacts of micro (nano) plastics in the aquatic and terrestrial environment. J. Hazard. Mater. 443, 130274.
Kankanige, D., Babel, S., 2020. Smaller-sized micro-plastics (MPs) contamination in single-use PET-bottled water in Thailand. Sci. Total Environ. 717, 137232.
Kirstein, I.V., Hensel, F., Gomiero, A., Iordachescu, L., Vianello, A., Wittgren, H.B., Vollertsen, J., 2021. Drinking plastics? - Quantification and qualification of microplastics in drinking water distribution systems by microFTIR and Py-GCMS. Water Res. 188, 116519.
Koelmans, A.A., Mohamed Nor, N.H., Hermsen, E., Kooi, M., Mintenig, S.M., De France, J., 2019. Microplastics in freshwaters and drinking water: critical review and assessment of data quality. Water Res. 155, 410–422.
Kumar, R., Manna, C., Padha, S., Verma, A., Sharma, P., Dhar, A., Ghosh, A., Bhattacharya, P., 2022. Micro (nano) plastics pollution and human health: how plastics can induce carcinogenesis to humans? Chemosphere 298, 134267.
Leban, G., Zupan, B., Vidmar, G., Bratko, I., 2006. Vizrank: data visualization guided by machine learning. Data Min. Knowl. Discov. 13, 119–136.
MacLeod, M., Arp, H.P.H., Tekman, M.B., Jahnke, A., 2021. The global threat from plastic pollution. Science 373 (6550), 61–65.
Makhdoumi, P., Amin, A.A., Karimi, H., Pirsaheb, M., Kim, H., Hossini, H., 2021. Occurrence of microplastic particles in the most popular Iranian bottled mineral water brands and an assessment of human exposure. J. Water Process Eng. 39, 101708.
Mintenig, S.M., Löder, M.G.J., Gerdts, G., 2014. Mikroplastik in Trinkwasser - Untersuchung Im Trinkwasserversorgungsgebiet des Oldenburgisch - Ostfriesischen Wasserverbandes (OOWV) in Niedersachsen. Probenanalyse Mittels Mikro-FTIR Spektroskopie. Oldenburgisch- Ostfriesischer Wasserverband (OOWV).
Mintenig, S.M., Löder, M.G.J., Primpke, S., Gerdts, G., 2019. Low numbers of microplastics detected in drinking water from ground water sources. Sci. Total Environ. 648, 631–635.
NHS 2021 Water, Drinks and Your Health, NHS. Online: https://www.nhs.uk/live-well/eat-well/food-guidelines-and-food-labels/water-drinks-nutrition/(Accessed 23/07/2023).
Ossmann, B.E., Sarau, G., Holtmannspotter, H., Pischetsrieder, M., Christiansen, S.H., Dicke, W., 2018. Small-sized microplastics and pigmented particles in bottled mineral water. Water Res. 141, 307–316.
Rubio, L., Marcos, R., Hernández, A., 2020. Potential adverse health effects of ingested micro-and nanoplastics on humans. Lessons learned from in vivo and in vitro mammalian models. J. Toxicol. Environ. Health, Part B 23 (2), 51–68.
Samandra, S., Mescall, O.J., Plaisted, K., Symons, B., Xie, S., Ellis, A.V., Clarke, B.O., 2022. Assessing exposure of the Australian population to microplastics through bottled water consumption. Sci. Total Environ. 837, 155329.
Saunders, K.J., 2012. Organic Polymer Chemistry: An Introduction to the Organic Chemistry of Adhesives, Fibres, Paints, Plastics and Rubbers. Springer Science & Business Media.
Scanlon, B.R., Fakhreddine, S., Reedy, R.C., Yang, Q., Malito, J.G., 2022. Drivers of spatiotemporal variability in drinking water quality in the United States. Environ. Sci. Technol. 56 (18), 12965–12974.
Schymanski, D., Goldbeck, C., Humpf, H.U., Furst, P., 2018. Analysis of microplastics in water by micro-Raman spectroscopy: release of plastic particles from different packaging into mineral water. Water Res. 129, 154–162.
Schymanski, D., Oßmann, B.E., Benismail, N., Boukerma, K., Dallmann, G., Von der Esch, E., Fischer, D., Fischer, F., Gilliland, D., Glas, K., 2021. Analysis of microplastics in drinking water and other clean water samples with micro-Raman and micro-infrared spectroscopy: minimum requirements and best practice guidelines. Anal. Bioanal. Chem. 413 (24), 5969–5994.
Shruti, V., Pérez-Guevara, F., Elizalde-Martínez, I., Kutralam-Muniasamy, G., 2020. First study of its kind on the microplastic contamination of soft drinks, cold tea and energy drinks-Future research and environmental considerations. Sci. Total Environ. 726, 138580.
Skjevrak, I., Due, A., Gjerstad, K.O., Herikstad, H., 2003. Volatile organic components migrating from plastic pipes (HDPE, PEX and PVC) into drinking water. Water Res. 37 (8), 1912–1920.
Statista 2023 Plastic production forecast worldwide 2025-2050. Online: https://www.statista.com/statistics/664906/plastics-production-volume-forecast-worldwide/(Accessed 23/07/2023).
Thompson, R.C., Moore, C.J., vom Saal, F.S., Swan, S.H., 2009. Plastics, the environment and human health: current consensus and future trends. Philos. Trans. R. Soc. B 364 (1526), 2153–2166.
Toplak, M., Read, S.T., Sandt, C., Borondics, F., 2021. Quasar: easy machine learning for biospectroscopy. Cells 10 (9), 2300.
Walker, T.R., Fequet, L., 2023. Current trends of unsustainable plastic production and micro (nano) plastic pollution. TrAC Trends Anal. Chem., 116984
Zhang, X., Lin, T., Wang, X., 2022. Investigation of microplastics release behavior from ozone-exposed plastic pipe materials. Environ. Pollut. 296, 118758.