Identification of Microplastics in Conventional Drinking Water Treatment Plants in Tehran, Iran

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Research Article

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Abstract

The presence of microplastics (MPs), as an emerging pollutant is a growing concern in different water resources. These particles are recognized as less than 5 mm in size. Most of the studies have been carried out in surface waters and wastewater treatment plants (WWTPs), but there are few studies on MPs in drinking water treatment plants (DWTPs). This study investigates these particles in three different conventional DWTPs in the city of Tehran, Iran and aims to analyze these particles down to the size of 1 µm. A scanning electron microscope (SEM) was utilized in this study to quantitatively analyze MPs. Accordingly, the average abundance of MPs in raw and treated water samples varied from 1996±268 to 2808±80 MPs L^{-1} and 971±103 to 1401±86 MPs L^{-1}, respectively. While particles smaller than 10 µm comprised 65-87% of MPs. Moreover, µ-Raman spectroscopy was used to characterize MPs. As the results, polypropylene (PP), polyethylene terephthalate (PET), and polyethylene (PE) were the most abundant identified polymers among MPs comprising more than 53% of particles. Additionally, MPs were categorized as fibers, fragments and spheres. This study fills the knowledge gap of MPs presence in Tehran DWTPs which is of high importance since they supply drinking water for more than 8 million people and investigates the performance of conventional DWTPs in removing MPs.

Introduction

Today, plastic products are found in almost all areas of our modern life such as clothing, cosmetics and health care, transportation, communication and food packaging to name a few (Huppertsberg and Knepper, 2018). Hence, the amount of plastic manufactured on a global scale reached 370 million metric tons in 2019 (PlasticEurope, 2020). Thereupon, trends of global plastic production, consumer-use patterns, inappropriate disposal of plastic wastes and demographics suggest an increase in plastic use in the future (Karbalaei et al. 2018). Since plastics hardly decompose due to the material properties, they remain in the environment for a long time (Sighicelli et al. 2018) and are a potential hazard for the environment due to their ubiquitous presence (Luo et al. 2018). This suggests an urgent need to investigate the risks which plastic particles might pose to living organisms and human beings. Identified as less than 5 mm in diameter, these small plastic particles or fibers are generally termed microplastics (MPs; Polanco et al. 2020). MPs are divided into two categories: primary and secondary. Primary MPs are specifically realized for various applications and can be in the form of exfoliating products, air-blasting technology and so forth (Cole et al. 2011). Secondary MPs, meanwhile, derive from the fragmentation of the larger items by weathering (Arthur and Baker 2009) and photodegradation (Barnes et al. 2009). Many studies have conducted research on MPs in marine environments (Sul and Costa 2014; Dobaradaran et al. 2018; Haan et al. 2019) and freshwater bodies (Blettler et al. 2017; Anderson et al. 2017; Ballent et al. 2016; Eriksen et al. 2013). Mao et al. (2020), for instance, investigated MPs in Wuliangsuhai Lake in northern China and detected these particles in all of the 32 samples with polystyrene and fibers as the most abundant polymer type and shape of MPs, respectively. The primary environmental risk associated with MPs is their bioavailability to marine organisms (Li et al. 2016) and ingestion by a large variety of farm and wild species (Miranda and Carvalho-Suzo 2016) in the shape of bioaccumulation. When body

ingests MPs, they absorb, distribute through the circulatory system, enter into different tissues, potentially resulting in several types of adverse effects (Foley et al. 2018; Chae and An 2017; Avio et al. 2017; Barboza et al. 2018). Most importantly, oxidative stress (Schirinzi et al. 2017), cytotoxicity (Anbymani and Kakkar 2018) and translocation to other tissues (Fiorentino et al. 2015). Likewise, the negative impact of distinct polymers on human health have been investigated. For instance, Polystyrene (PS) and Polyvinyl chloride (PVC), trigger reproductive abnormalities (Wang et al. 2016), inflammatory gene expression and cell morphology of human gastric adenocarcinoma epithelial cells (Forte et al. 2016). The number of studies evaluating MPs in water bodies are numerous throughout the world, however, limited studies have investigated the presence of MPs in WWTPs, DWTPs and groundwater resources (Mintenig et al. 2019; Magni et al. 2019; Wolff et al. 2018; Sun et al. 2019). Thus, further research on these facilities is of great need. For example, Pivokonsky et al. (2020) investigated two DWTPs of the same river and they detected MPs ranging from 20 to 1200 particles in each L of their samples, with fragments as the most abundant shape of MPs larger than 1µm. Since DWTPs are home to the purification of water resources to millions of people in cities, the purpose of this study is to evaluate the presence and polymer types of MPs in Tehran DWTPs and investigate the amount of MPs removal in the outlet of DWTPs. To the knowledge of the author, this is the first study that aims to evaluate MPs and the reduction efficiency of DWTPs in Iran.

Materials And Methods

Sampling site and sample collection

In total, there are five conventional DWTPs in Tehran with the same system of water purification, including screening, coagulation and flocculation, sand filtration and disinfection that three of them are fed by different rivers, so these three DWTPs were chosen to be investigated. In this study, the names of DWTPs were not mentioned for confidentiality reasons, so they are named DWTP 1 to 3. The DWTP number 1 is fed from Karaj and Kan River in the northwest of Tehran with the capacity of 7.2 m^3 s^{-1}, the source of the second DWTP is the Jajrud river in the northeast of Tehran with a capacity of 4 m^3 s^{-1} and the third DWTP receives water from Lar Dam in the northeast of Tehran with the capacity of 5.7 m^3 s^{-1}. For the sampling, dark glass bottles with a capacity of 2.5 L were used to collect water samples from the raw and treated water from the DWTPs. Sampling was conducted in three different time intervals, over five months from April to September, from 12 am to 1 pm. Each time, one sample from raw water and one sample from treated water were taken (18 samples comprising 45 L in total). Subsequently, collected samples were kept in the dark at 4 ºC before sample preparation.

Sample preparation

To prepare the samples, the Wet Peroxide Oxidation method (NOAA, 2015) was applied to digest microorganisms. To explain briefly, a 0.05 M Fe (II) solution was prepared by 15 g $FeSO_4.H_2O$ and 6 mL concentrated sulfuric acid (Merck Millipore, USA) and one L of deionized water. Every L of samples was added 80 mL of Fe (II) solution and 80 mL 35% hydrogen peroxide (Merck Millipore, USA) (Wang et al.

2020; Pivokonsky et al. 2018; Anderson et al. 2017; Masura et al. 2015). Afterward, One L Erlenmeyer flasks containing samples were placed on a stirring plate at 60 ºC at 300 rpm for 30 minutes to boost the digestion. Subsequently, samples were kept at room temperature for 24 h before filtration. Then they were filtered via a vacuum pump through cellulose nitrate membrane filters with a pore size of 0.2 µm and 47 mm diameter. To remove clay and other inorganic particles from the filters, a density separation was applied. Hence, a zinc chloride solution (Merck Millipore, 5 M, 1.55 g cm^{-3}) (Yang et al. 2019; Mintenig et al. 2019; Quinn et al. 2017) was added to 20 mL centrifuge tubes and the filters were placed into the solution. Subsequently, the tubes were treated with an ultrasonic bath for 10 minutes to detach the particles from the filter. Without removing the filters, the tubes were centrifuged at 4000 rpm for 5 minutes and supernatants were filtered again to have almost pure MPs. Afterward, filters were placed in Petri dishes and dried in an oven at 60 ºC for 1 h. Afterward, the Petri dishes containing the samples were covered by aluminum foil and placed in a desiccator for further quantitative and qualitative analysis.

Quality assurance/Quality control (QA/QC)

Cotton laboratory outfit and nitrile gloves were utilized to minimize the risk of pollution. A negative-pressure ventilation system was functioning during the sample processing to eliminate the risk of depositing airborne MPs on the filters. Working surfaces on which the experiments were conducted were repeatedly cleaned with 1 M NaOH (Merck Millipore, USA). Furthermore, all the glassware for the sample processing were rinsed three times with filtered deionized water to remove potential MPs on their surfaces. For the sampling, dark glass bottles were used to lower the effect of photo-degradation. Plastic bottles were abstained from using in order to minimize the risk of the addition of MPs from the bottles. To minimize sample pollution, a layer of aluminum foil was placed between the bottles and screw caps. Additionally, one blank sample was carried out to ensure if there was sample contamination. The preparation method was applied on one L of previously filtered deionized water.

Quantitative analysis

A scanning electron microscope (Thermo Fisher Scientific, FEI Quanta 200, USA) with an accelerating voltage of 30 kV and detector working distance of 10 mm was used to image the filter surface in order to enumerate and identify the shape and size of MPs. The filters were cut in half and three cut-outs (3 mm ×8 mm) from one of the halves (one in the center, one in the edge and one in the middle) were scanned by SEM (Pivokonsky et al. 2018). Before imaging, a gold layer was sputtered onto the samples to create electrical conductivity. Approximately 60 images from each cut-out were taken (13 mm^2 in total) and the number, size and shape of detected MPs were extrapolated to the whole area of the filter. The number, size and shape (fiber, fragment and sphere) of MPs were verified by ImageJ software (Version 1.50e, National Institute of Health, USA) based on one-L samples. Fibers were verified as thin and long MPs and were measured by their thickness, while fragments are particles that have been created by breaking down of larger pieces of plastic via degradation that were measured by their longest end and spherical MPs

have an appearance of a sphere. The particles were divided into 5 categories in terms of their size (1-5 µm; 5-10 µm; 10-50 µm; 50-100 µm; >100 µm).

Qualitative analysis

To identify the chemical properties of MPs, a previously cleaned needle was used to transfer MPs on the another half of the filter onto a conductive adhesive copper tape using a light microscope (N-120, Hinotek, China). In total, 107 suspected MP particles with various shapes and sizes were carefully transferred onto the copper tape and sent for analysis. Since Fourier Transform Infrared (FTIR) spectroscopy is used for detecting the spectrum of particles greater than 500 µm and µ-FTIR is utilized for the particles down to the size of 20 µm (Li et al. 2018) and the fact that the majority of the MPs in this study were comprised of particles smaller than 20 µm, µ-Raman spectroscopy (Horiba Scientific, XploRA ONE^{TM}, Japan) was used to detect the chemical composition of MPs by Labspecs 6 Software (Horiba Scientific, Japan) and the obtained spectra were compared with Infrared and Raman Users Group (IRUG). The frequency of excitation laser of micro-Raman spectroscopy was 785 nm, 1 accumulation and 100× objective at 10 mW. The grating was 1200 1 mm^{-1}, instrument aperture 100-µm slit, acquisition time 2 s and spectral range was set to 500-3500 cm^{-1}. In qualitative analysis, 62% of suspected particles were MPs, so this amount was subtracted from the MP numbers in quantitative analysis.

Statistical analysis

All statistical analyses were computed using Statistical Package for Social Science (version 16.0, SPSS, Inc.) and the figures were created with Microsoft Excel 2016 for Windows. Prior to statistical analysis, all data were tested for the basic assumptions for normality and homogeneity of variance. The Kolmogorov-Smirnov test was applied to analyze the normality of the data distribution. $P < 0.05$ was considered statistically significant (Tables S1-S3, supplementary data).

Results And Discussion

MP abundance and size

Three DWTPs are fed from three different rivers and there are no industrial areas in the vicinity of them. However, these rivers pass through the city of Tehran, so MP pollution is expected to be originated by people activity (Schmidt et al. 2020) and deposition of airborne MPs (Dehghani et al. 2017). Therefore, in all of the samples, both in raw water and treated water of DWTPs, MPs were detected in various numbers. The average number of MPs in raw water of DWTPs are 2808±80, 1996±268 and 2172±119 particles L^{-1} in DWTP 1, DWTP 2 and DWTP 3, respectively. The difference in the number of MPs can be attributed to the variation in water quality, precipitation and density of population in the proximity of the rivers. Kankanige and Babel (2020) investigated a conventional DWTP in Thailand for MP removal and they

reported that raw water samples of rainy seasons are 30% more polluted with MPs than in dry seasons. Moreover, source of water supply may influence MP abundance. For instance, Jajrud River (source of DWTP 1) originates from Latyan Dam, where is a hotspot for people's recreation. While Lar dam (source of DWTP 3) is almost free of people's activity and water is transferred by a tunnel with a diameter of 3.6 m to the DWTP 3. However, a detailed study needs to be conducted to evaluate the relation between quality of water and abundance of MPs. The quality of raw water in this study is described in Table S4, supplementary data.

Regarding treated water samples, the average number of MPs are 1401±86, 1042±269 and 971±103 particles L^{-1} for DWTP 1, DWTP 2 and DWTP 3, respectively. The function of all three DWTPs is the same with the same coagulant –Ferric chloride– and the same purification system. Table 1 represents the number of MPs in both raw and treated water samples. Failure in desirable removal of MPs can be observed in all the DWTPs – an average decrease of 50.1, 48.4 and 55.2 in DWTP 1, DWTP 2, and DWTP 3, respectively. The rate of removal in this study is comparable to that of Kankanige and Babel (2020) with 57.2% and 67.6% in rainy and dry seasons, respectively. Lower removal rate in this study can also be attributed to lower efficiency of DWTPs. For example, Polyacrylamide (PAM) is not used in the coagulation process. However, Pivokonsky et al. (2018) indicated that more than 20% of MPs in their WTP 1 are comprised of PAM due to its usage in coagulation process. They also demonstrated that two WTPs that operate granular activated carbon (GAC) filtration, remove more than 80% of MPs, while the other one without this filtration system can remove 70% of these particles. In this study, performance of three DWTPs in the reduction of MPs is almost the same which can be attributed to the same functionality of them. According to fig. 1, 65% to 87% of MPs are comprised of particles smaller than 10 µm. This implies that the findings in this study are comparable to other similar studies investigating MPs in drinking and bottled water (Tong et al. 2020; Wang Z. et al. 2019; Schymanski et al. 2018; Pivokonsky et al. 2018). Table 2 compare the result of this study to that of other similar studies. Although the diminishment of larger MP particles was significant, most of the smaller MPs were not removed due to lower density, the failure in coagulation and flocculation process and sand filtration. However, almost all of the particles greater than 50 µm were removed in the treatment process. Additionally, a significant portion of MPs are smaller than 1 µm, but due to the difficulty in the enumeration process and the fact that those particles cannot be analyzed qualitatively, they were disregarded in quantitative analysis.

Table 1

Number of MPs in each sample (L^{-1}). Removal rate of each sampling day was calculated by the difference in the abundance of MPs in water and treated water (L^{-1}). The average number of MPs in each DWTP is recorded above.

Removal (%)	MPs in treated water (L^{-1})	MPs in raw water (L^{-1})	
			DWTP 1
51.8	1312	2725	Day 1
50.0	1407	2814	Day 2
48.5	1484	2884	Day 3
50.1	1401±86	2808±80	Average
			DWTP 2
53.2	895	1911	Day 1
41.2	1352	2297	Day 2
50.7	878	1781	Day 3
48.4	1042±269	1996±268	Average
			DWTP 3
56.4	891	2045	Day 1
59.0	934	2280	Day 2
50.3	1088	2191	Day 3
55.2	971±103	2172±119	Average

Table 2
Comparison of other similar studies to the findings of this study

Reference	MP abundance (L^{-1})		Size range of MP particles (µm)	Type of Water
	Range	Mean		
Pivokonsky et al. (2018)	1383–1575	1473±34	>1	WTP 1, raw water
Pivokonsky et al. (2018)	1648–2040	1812±35	>1	WTP 2, raw water
Pivokonsky et al. (2018)	3123–4464	3605±497	>1	WTP 3, raw water
Pivokonsky et al. (2018)	369–485	443±10	>1	WTP 1, treated water
Pivokonsky et al. (2018)	243–466	338±76	>1	WTP 2, treated water
Pivokonsky et al. (2018)	562–684	628±28	>1	WTP 3, treated water
Tong et al. (2020)	0-1247	440	>1	38 tap water samples
Wang Z. et al. (2019)	930 ± 72	6614 ± 1132	>1	ADWTP [a]
Simon et al. (2018)	2223-18285	7216*	10-500	Influent of 10 WWTPs
Simon et al (2018)	19-447	54*	10-500	Effluent of 10 WWTPs
Kankanige and Babel (2020)	-	1590.8 ± 148.8	>6.5	Influent of a conventional WTP
Kankanige and Babel (2020)	-	609.1 ± 84.7	>6.5	Effluent of a conventional WTP
This study	1867-2665	2255±383	>1	DWTP 1, raw water
This study	1434-1699	1588±313	>1	DWTP 2, raw water
This study	1899-1966	1933±381	>1	DWTP 3, raw water
This study	1201-1600	1356±264	>1	DWTP 1, treated water
This study	900-1167	1022±259	>1	DWTP 2, treated water
This study	1133-1368	1222±288	>1	DWTP 3, treated water

*Median

MPs shapes

Regarding the shape of MPs, fibers outnumbered the other two categories with 51.1% in raw water samples, while fragments comprised 35.6% of the MPs. However, fibers were removed higher in comparison with fragments, comprising 38% of MPs in treated water samples. That may be attributed to their long ends which can be trapped in sand filtration process. These microfibers can be originated from washing fabric garments (De Falco et al., 2019). Moreover, fragments were more abundant in treated water than in raw water. These particles are created through the breakdown of larger plastic debris and plastic products including packaging materials, plastic bottles and washing synthetic materials (Wang S. et al. 2020; Wang Z. et al. 2019; Auta et al. 2017). Fig. 2 represents the different shapes of MPs detected in samples. Finally, spherical MPs were the least abundant among detected MPs (13.3% and 5.3% in raw water and treated water samples, respectively). These particles originate from personal care and cosmetic products (Wang Z. et al. 2020). Wang Z. et al. (2020) also demonstrated that sand filtration is able to remove 60-80% of microspheres which is comparable to that of this study (60.2%). Fig. 3 illustrates three different shape of detected MPs in both raw and treated water samples.

Qualitative analysis

In qualitative analysis of MPs, 10 different polymers were detected by μ-Raman spectroscopy (PP, Polyethylene Terephthalate (PET), PE, Polystyrene (PS), Polytetrafluoroethylene (PTFE), Polyurethane (PU), Polyamide (PA), Polybutylene terephthalate (PBT), Polyvinylidene Fluoride (PVDF), Polyvinyl Chloride (PVC) and polycarbonate (PC)). Fig. 4 represents the average composition of MPs in raw and treated water samples. PP was the most abundant polymer in both raw and treated water samples with 27.3% and 24.8%, respectively. The high presence of these polymers can be attributed to the widespread consumption of food packaging materials, including cereal, flour, biscuits, dried fruit and vegetables, dried pasta (Vera et al., 2020). PET is the second common polymer detected by μ-Raman spectroscopy with 15.1% in raw water samples. However, it is the third common polymer in treated water samples (14.2%), following PE with 12.1% and 14.4% of raw and treated water samples, respectively. PET is also consumed considerably as beverage bottles and bottled water (Kang et al. 2016), PE is usually used for bags, packaging and houseware (Pivokonsky et al. 2020). The result from qualitative analysis in this study is consistent with other similar studies. For instance, Tong et al. (2020) demonstrated that 26.7% and 24.4% of MPs from tap water in China are PE and PP, respectively. Wang Z. et al. (2020), by investigating an advanced drinking water treatment plant (ADWTP) in China, showed that 55.4-63.1%, 15.1-23.8% and 8.4-18.2% of MPs in raw water samples are comprised of PET, PE and PP, respectively. Likewise, they indicated that PAM, prior to PET (47.2-58.8%), is the most abundant MPs in treated water samples, comprising 10.1-14.7% of MPs.

Further study

In this study, we demonstrated that treated water from conventional DWTPs contains a significant amount of MPs, so people are exposed to MP ingestion from drinking water. Mortensen (2021) also indicated that drinking water is one of the main sources of ingestion of MP smaller than 50 μm. PP, PET and PE MPs that are the most abundant in treated water samples, are recognized to pose human health risks (Çobanoğlu et al. 2021; Stock et al. 2021). These particles can also adsorb and transfer chemical to humans (Abbasi et al. 2021; Dong et al. 2020). Therefore, further studies are recommended to boost the efficiency of DWTPs to remove MP particles. In this regard, addition of GAC filtration needs to be investigated to increase MPs removal. Increasing depth of sand filtration and using finer particles in these systems can also trap more MPs, especially microfibers. Usage of PAM which can better remove MPs in coagulation/flocculation process is recommended. However, it can be a source of pollution in treated water, so an alternative coagulant aid is suggested. Moreover, sources for DWTPs water supply can be managed to be free of human activity and transferring water in a tunnel can lower MP pollution of raw water. Additionally, plastic pipes are better to be disregarded for water transfer—whether from source to DWTP or DWTP to household consumption—or be checked up regularly in case of potential degradation.

Conclusion

This study investigated MPs in three different conventional DWTPs of Tehran through quantitative and qualitative analysis. MPs were abundant both in raw and treated water samples. On average, 1996±268 to 2808±80 MPs L^{-1} and 971±103 to 1401±86 MPs L^{-1} were identified in raw and treated water samples, respectively. Accordingly, conventional DWTPs are not successful to efficiently eliminate MPs. Moreover, 65-87% of MPs are smaller than 10 μm that were more abundant in treated water than in raw water which indicates that conventional DWTPs are incapable of removing MPs in this size. Generally, the ability of MPs removal by the investigated DWTPs ranges from 41.2% to 59.0%. Additionally, PP was the most abundant type of MPs in three detected DWTPs, both in raw and treated water samples, comprising 27.3% and 24.8%, respectively. Furthermore, fibers were more abundant than fragments and spheres in raw water, (51.1%), while in treated water, fragments were more abundant than other two categories (56.7%).

Declarations

1. Funding

All the expenditures on the findings of this study were provided by the authors.

2. Competing interest

This manuscript has not been submitted to, nor is under review at, another journal or other publishing venue. The following authors have affiliations with organizations with no financial support in the subject matter discussed in the manuscript.

3. Availability of data and material

Not applicable

4. Ethics approval and consent to participate

Not applicable

5. Consent for publication

Not applicable

6. Consent to participate

Not applicable

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8. Author contribution

DA conducted laboratory experiments and preparing literature. RM assisted in writing the literature and revision. HT did the sampling and prepared statistical data.

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Figures

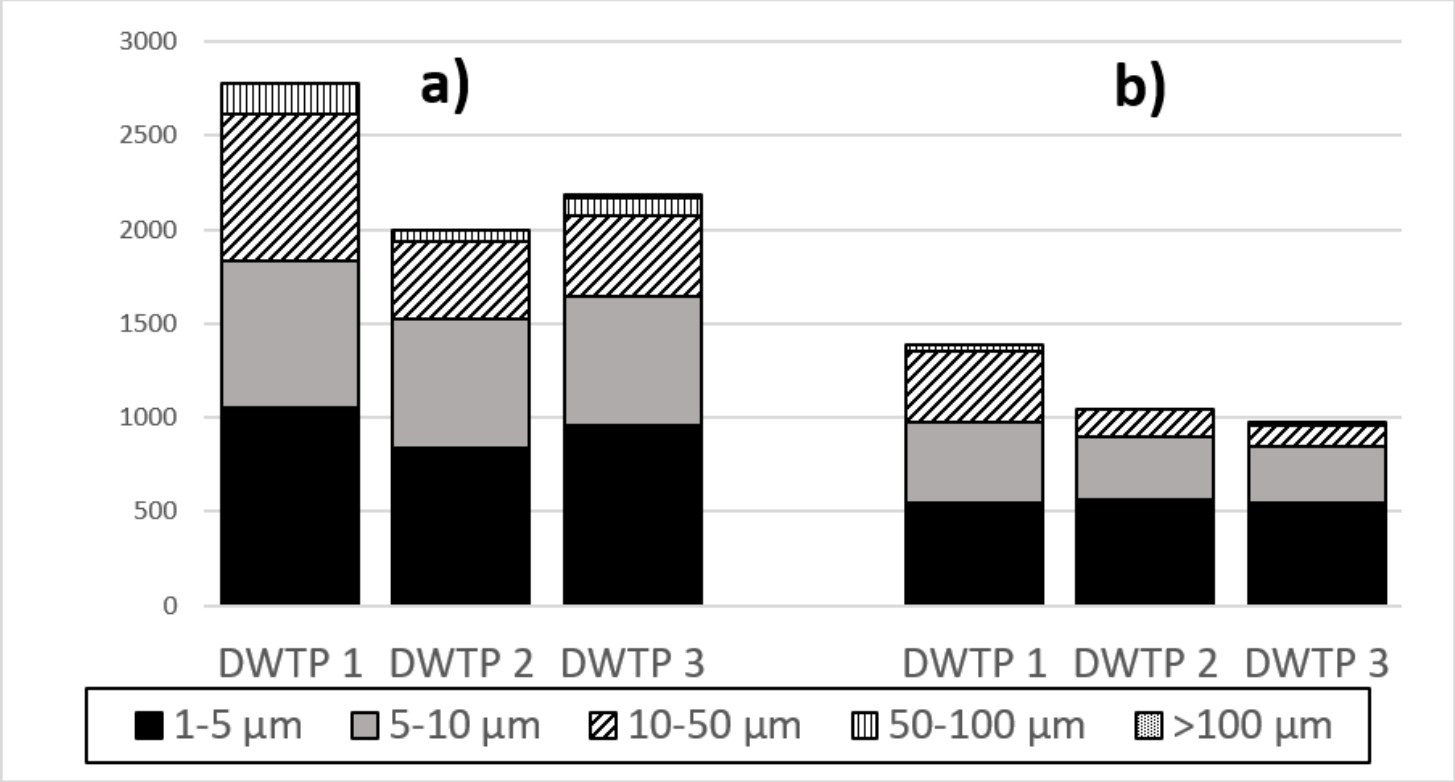

Figure 1

Average size distribution and number of MPs detected in each L of (a) raw water samples and (b) treated water samples

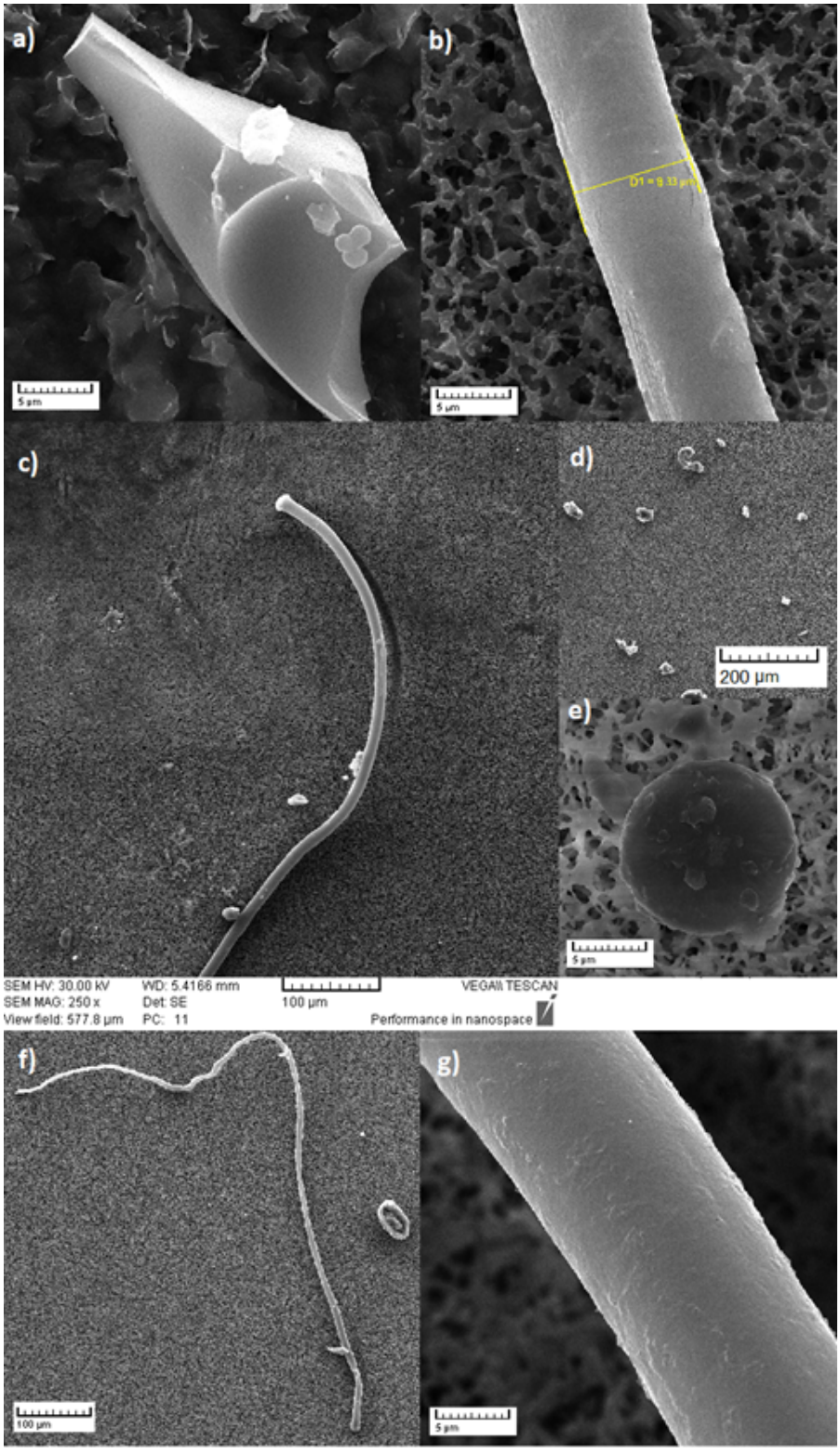

Figure 2

Images (a) and (d) depicts fragments in various sizes, figures (b), (c), (f) and (g) illustrate fibers in different diameters and (e) figure represents an aged microsphere

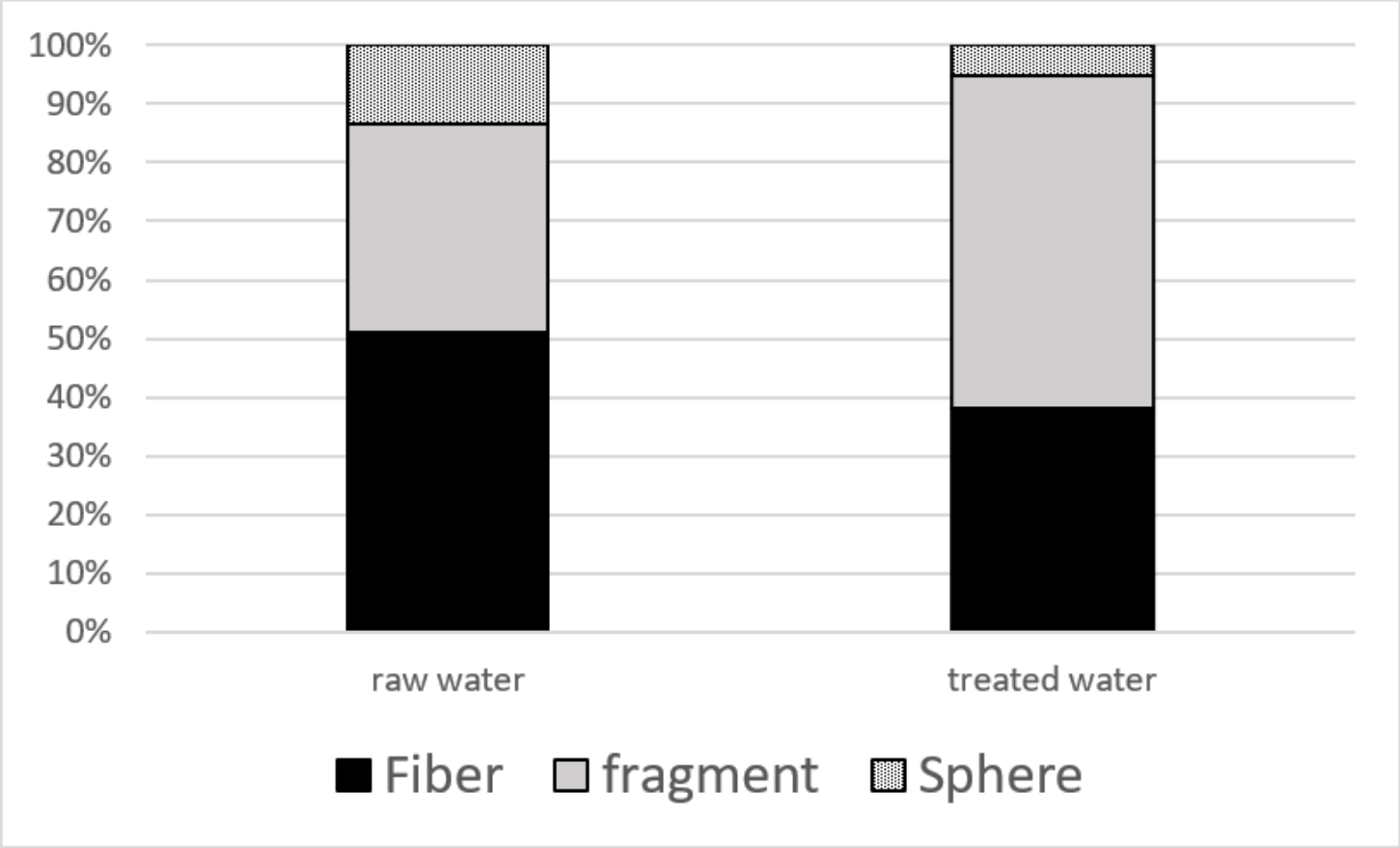

Figure 3

Abundance of MPs in three different shapes in raw and treated water

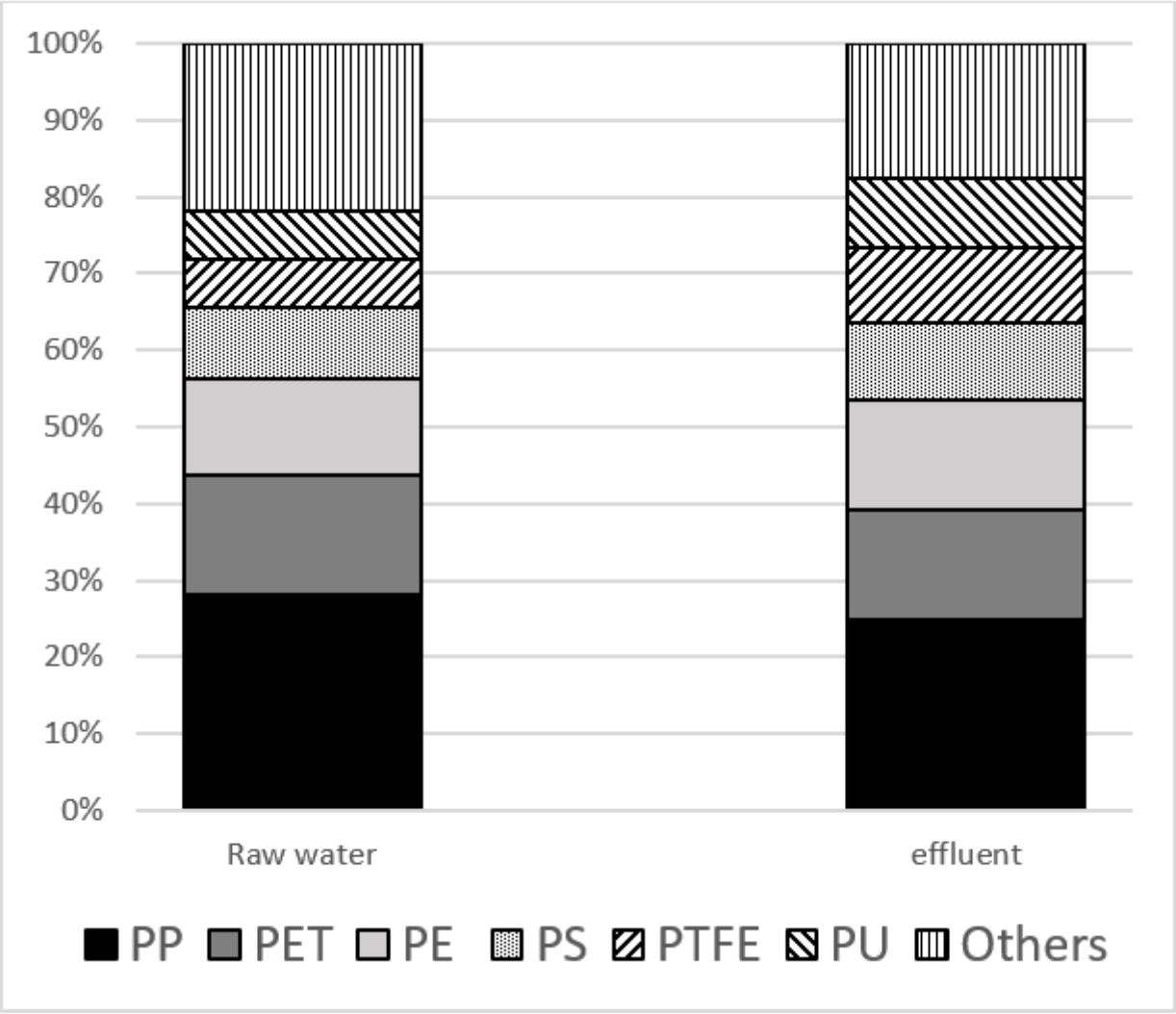

Figure 4

Average composition of MPs detected by μ-Raman spectroscopy in both raw water and treated water samples.

Supplementary Files

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