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Identification, Quantification, and Evaluation of Microplastics Removal Efficiency in a Water Treatment Plant (A Case Study in Iran)

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ABSTRACT: Microplastics (MPs) are among the emerging pollutants that recently attracted the researcher's attention around the world. These particles can absorb other chemicals, and microbial contaminants and enter them into the food chain, and environment. This study was conducted to investigate the occurrence of MPs in raw and treated drinking water and evaluate the MPs removal efficiency in a drinking water treatment plant (DWTP) in Iran. MPs particles were counted at different stages of DWTP, using a scanning electron microscope after the initial preparation steps include filtration, and chemical digestion, and then examined for the nature of the particles using a micro-Raman spectrometer. The concentration of MPs in influent, clarifier's effluent, and DWTP's effluent were 1597.7 ± 270.3, 676.2 ± 69.0, and 260.5 ± 48.9 MPs/L, respectively. The total efficiency of the DWTP in MPs removal was 83.7%, which the clarification and filtration stage removed 57.7%, and 26.0% of total MPs, respectively. The most abundant polymers identified were PP, PE, and PET. Despite the effective removal of MPs in the DWTP, on average $2.25 \times 10^{11} \pm 4.23 \times 1010$ MPs are daily discharged into the water distribution system through the effluent of this DWTP.

KEYWORDS: Microplastic, plastic, drinking water treatment plant, removal efficiency, drinking water

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Background

The microplastics (MPs) are defined as synthetic plastic particles less than 5 mm and more than 1 µm in diameter (Crawford & Quinn, 2016; Ranatunga et al., 2021.). The world first became aware of MPs in the ocean in 1972, and since then many studies have investigated this emerging threat (Crawford & Quinn, 2016.). The entry of MPs into the environment has increased in recent decades due to the increasing use and production of plastics, and this emerging pollutant is now commonly found everywhere in the environment (Carr et al., 2016; Sharifi & Movahedian Attar, 2021b). In 2018, approximately 360 million tons of plastic were manufactured in the world, and the annual growth rate of plastic production in the period 1950 to 2015 was 8%. It is also estimated that future production will reach to 1.1 billion tons by 2050 (Zhang et al., 2022). Therefore, MPs have become a global concern and have attracted the attention of researchers around the world in recent years. Since there is not enough information about the potential risk associated with MPs, the actual risk posed by MPs has not yet been determined (Xu, Peng et al., 2018). However, the MPs are able to carry microbial contaminants(Marsden et al., 2019) and absorb the drugs and persistent organic contaminants such as PBDEs[1], PAHs[2], and PCBs[3] (Carr et al., 2016; Sørensen et al., 2020; Xu, Hou et al., 2018). Under such conditions, the presence of MPs in food and water can expose humans to these contaminants (Lee et al., 2021; Makhdoumi et al., 2021). Also, due to the attractive coloration, small size, and length to diameter ratio of MPs, they are readily ingested by aquatic organisms and can enter into the food chain (Dris et al., 2018; Jovanović, 2017).

There is limited data about the MPs removal process in drinking water treatment plants (DWTP). Pivokonsky et al. (2018) studied raw and treated drinking water in three DWTPs in the Czech Republic for MPs and found 1,473 ± 34 to 3,605 ± 497 MPs/L and 338 ± 76to 628 ± 28 MPs/L in raw and treated drinking water, respectively (Pivokonsky et al., 2018). They found that 77.6% of MPs were removed in these DWTPs. Wang et al. (2020) examined an advanced DWTP in China for MPs and found that the raw and treated drinking water contained 6,614 ± 1,132 and 930 ± 72 MPs/L, respectively. Their study revealed that the DWTP remove MP with efficiencies ranging from 82.1% to 88.6%.(Wang et al., 2020). Sarkar et al. (2021) investigated the MPs in different stages of a DWTP with pulse clarifier in India and found that 17.83, 17.53, 17.11, 6.99, 11.17, and 2.75 MPs/L were present in raw water, pre-disinfection, flocculation, pulse clarification, sand filtration, and treated water, respectively. The total removal efficiency of MPs in their study was 85.39% (Sarkar et al., 2021).

As mentioned earlier, the presence of MPs in DWTPs has been proven in studies and it has been shown that the DWTPs can provide a barrier for direct discharge of MPs into the drinking water systems (Pivokonský et al., 2020), but there is still no treatment technology specifically designed for MPs removal, and there is no legislative limit for the presence of MPs in drinking water (Novotna et al., 2019). In addition, the World Health Organization report points out the importance of conducting more research on MP in different stages of DWTPs (Marsden et al., 2019). This study aims to investigate the concentration of MPs in different stages of a DWTP in Iran and to determine the MPs removal efficiency in different stages of this treatment plant.

Methods

The MPs measurement steps include sampling and filtration, pretreatment, counting, and quantitative and qualitative analyses is described below:

Sampling

In this study, a DWTP in Iran was investigated for MPs. The DWTP treats surface water with a capacity of 10,000 L/S, and its stages include coagulation/flocculation, Clarification, and sand filtration. The sampling of this DWTP was done six times in, October, November, and December 2020. The MPs concentration (MPs/L) was measured in different stages of the DWTP's include influent, clarifier's effluent, and DWTP's effluent. Each time, two 1 L of these steps were sampled and transferred to the lab with the glass containers.

Filtration and digestion

This step was performed following the study of Mintenig et al. (2019) and Wang et al. (2020) using the WPO (Wet Peroxidation Oxidation) method. First, the samples were passed through a cellulose nitrate filter (Sartorius, Cellulose Nitrate, 47 mm, 0.45 μm) using a vacuum set, and the filters were rinsed using distilled water to the clean 1 L beakers. Thirty milliliters of H_2O_2.35% (Dr. Mojallali, Iran) was then added to the samples and covered with a watch glass and placed in the oven at 40°C for 24 hours. Then, a series of samples was passed through a hydrophilic PTFE filter (FILTERBIO, PTFE, 0.47 mm, 0.45 μm) for quantitative analysis, and a series was passed through a fiberglass filter (Whatman, GF-3, 125 mm, 0.6 μm) for qualitative analysis using a glass vacuum set. The filters are then dried at room temperature and transferred to a clean glass Petri dish for the next analyses.

Qualitative analyses

UniRAM Raman spectrometer equipped by a solid-state laser with an excitation wavelength of 785 nm and a power of 200 mW, used for qualitative analyses (Frére et al., 2016; Ghosal et al., 2018). Two cut-outs (1 cm × 1 cm) of each filter were mounted on the Au-coated glass holder, and Raman spectra (surface-enhanced Raman spectroscopy) were recorded. Two spectra were recorded from each cut-out. Then the spectra were baseline corrected using Origin 2019 software and the spectra were compared to the reference spectra (Crawford & Quinn, 2016.) and the MPs type was identified.

Quantitative analyses

Four cut-outs (≈5 mm × 8 mm) from each PTFE filter were analyzed and photographed using a scanning electron microscope (SEM) (Philips XL30 ESEM, Netherlands). A layer of conductive gold was sputtered onto the filters prior to analysis (Sharifi & Movahedian Attar, 2021a). MPs were counted based on their size (<10, 10–50, 50–100, and >100 μm) and shape (fibers, fragments, ovals, and spheres). The exact size of MPs and filters was measured using SEM. Then, the MPs concentration in 1 L of each sample was calculated by comparing the cot-outs area with the total area of filters. In addition, the results of the quantitative analysis were modified based on the results of the qualitative analysis and the blank samples, and the percentage of particles identified as non-plastic and the MPs concentration in the blank samples were subtracted from these results.

MPs removal efficiency in each stage was calculated based on the difference of MPs concentration, before and after that stage and analysis of the overall efficiency of the treatment plant in MPs removal was calculated based on the difference of MPs concentration in the effluent and influent.

Controls

To increase the accuracy of the results, the following steps were performed based on the suggestion of Marsden et al. (2019) and Crawford and Quinn (2016).

- The results of each step were repeated six times in 3 months.
- Results were corrected based on the percentage of non-plastic materials and blank samples.
- The equipment used for sampling and analysis was made of glass. They were also washed with acid and rinsed three times with distilled water before use.
- The work surfaces were cleaned with 70% ethanol before each use.
- Air movement in the laboratory was controlled by closing the windows and analyzes were performed under the fume hood with laminar flow.

Statistical analysis

The mean values and SD of triplicate samples from different sampling days were calculated and expressed as abundance of MPs. All statistical analyses were calculated and graphed using Microsoft Excel (2017 version).

Results and Discussion

Characterization of MPs

Seventy-two spectra were obtained from the samples by micro-Raman and compared with the reference spectra. In addition, 16.7%, 8.3%, and 12.5% of the particles in the influent, clarifier's effluent, and effluent, respectively, had unknown spectra that were subtracted from the MPs concentration. The most frequently identified polymers were PP, PE, and PET, respectively. This was roughly consistent with the study by Pivokonsky et al. (2018). These polymers were more abundant than other polymers in other studies, which could be due to the high production and persistence of these polymers (Li et al., 2020). It should be mentioned that the inorganic and organic substances bound to MPs may alter the Raman spectra and these spectra

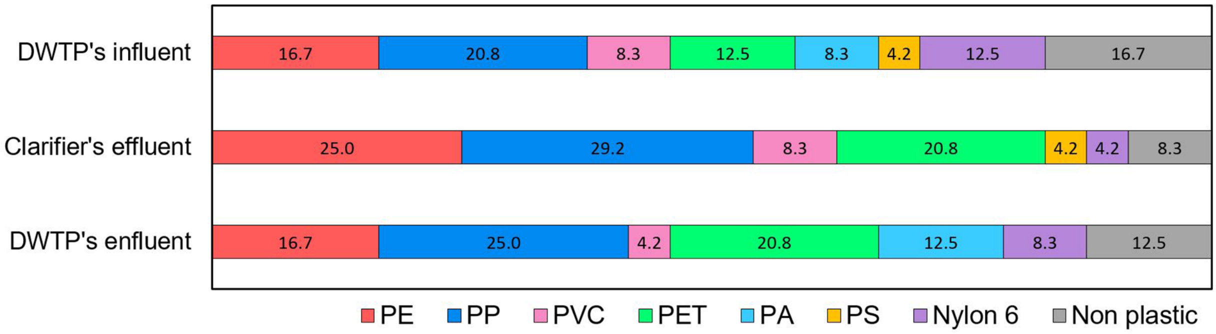

Figure 1. The abundance of polymers detected in the DWTP.

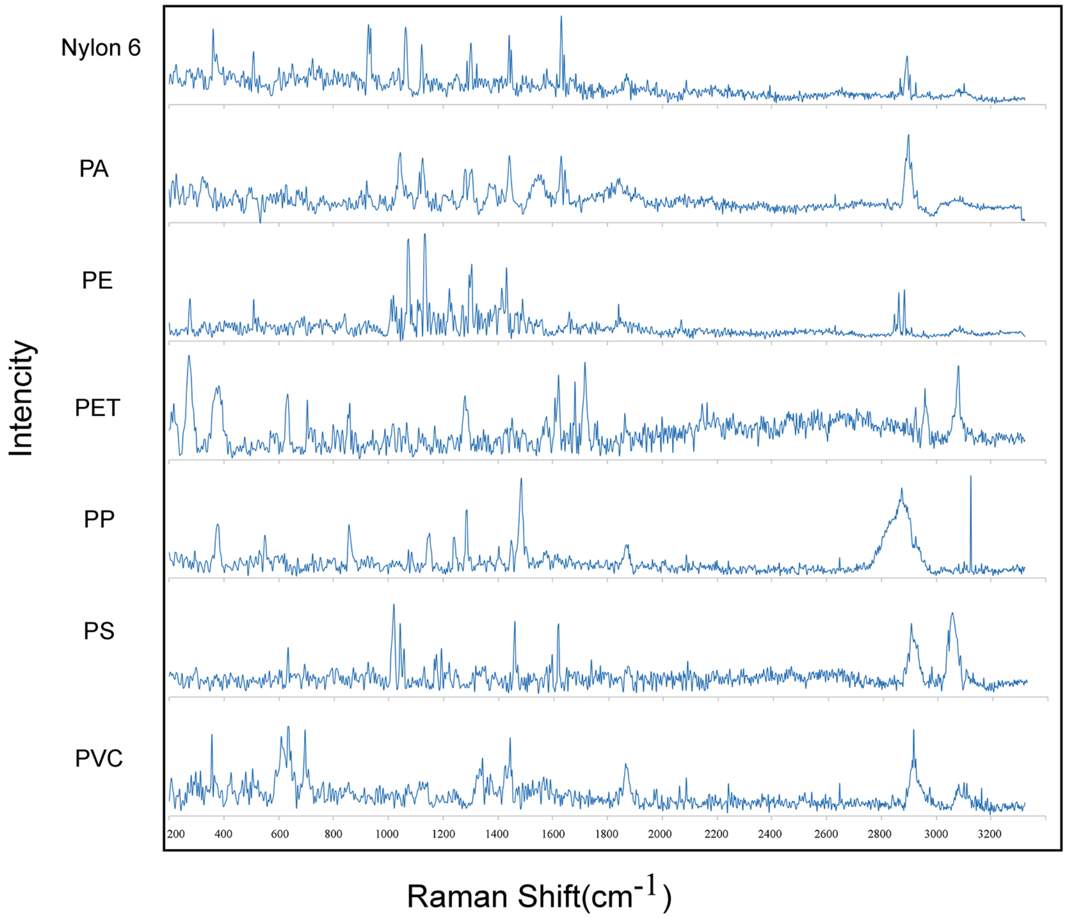

Figure 2. Identified polymer spectra.

may not completely match the reference spectra (Lenz et al., 2015).The abundance of polymers detected in the DWTP is shown in Figure 1, and the identified polymer spectra are shown in Figure 2.

Abundance of MPs

The mean and deviation of MPs in influent, after clarifier, and effluent were 1597.7 ± 270.3, 676.2 ± 69.0, and 260.5 ± 48.9 MPs/L, respectively. In this study, MPs were divided into four categories based on their size (<10, 10–50, 50–100, and >100 μm) and four categories based on their shape (fibers, fragments, ovals, and spheres). The percentage of MPs based on shapes and sizes in different stages of the DWTP is shown in Figure 3, and the microscopic images are shown in Figure 4. The fiber and fragment was the dominant shape and MPs ⩽ 10 μm was the dominant sizes in all sampling sites. The results related to the percentage of MPs based on shapes and sizes are strongly consistent with the result of (Pivokonsky et al., 2018). The concentrations of MPs in the influent and effluent of different DWTPs are listed in Table 1. As can be seen in this table, the MPs

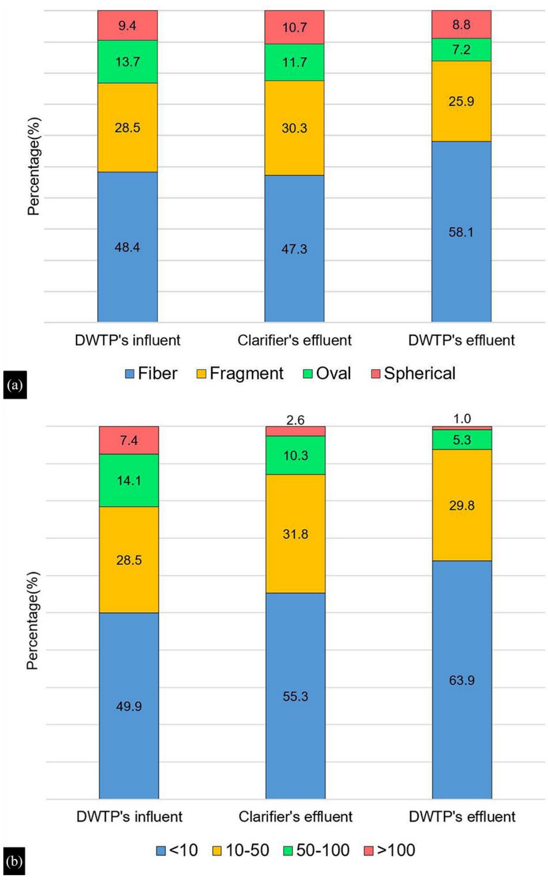

Figure 3. The percentage of microplastics based on sizes (a) and shapes (b) in different stages of DWTPs.

concentration in DWTPs influent has been varied from 17.88 particles per liter in the study of Sarkar et al. (2021) to 6614 ± 1132 particles per liter in the study of Wang et al. (2020). Also, the MPs concentration in DWTPs effluent has been varied from 2.75 particles per liter in the study of Sarkar et al. (2021) to 930 ± 71 particles per liter in the study of Wang et al. (2020).

MPs removal efficiency in DWTP

The MPs removal efficiency in the clarification and sand filtration stage were 57.7% and 61.5%, respectively. The MPs removal efficiency in the different stages of DWTP based on the shapes, sizes, and types of MPs is shown in Table 2. As can be seen in this table, the removal efficiency of MPs with a size more than 100 µm, oval shape, and PET type was higher than other MPs. The overall MPs removal efficiency of DWTP in MPs removal was 83.7%, which clarification and filtration stage removed 57.7% and 26.0% of total MPs, respectively. This is in strong agreement with the results of Sarkar et al. (2021). They found that a pulse clarification was able to remove 62% of MPs and 23% of them were eliminated by sand filtration. MPs removal efficiencies in DWTPs in different studies are shown in Table 1. As can be seen in this table, the MPs removal efficiencies in DWTPs, range from 40% to 88.6% in the study of Pivokonský et al. (2020) and Wang et al. (2020), respectively. Final treatment technologies such as ozonation, and

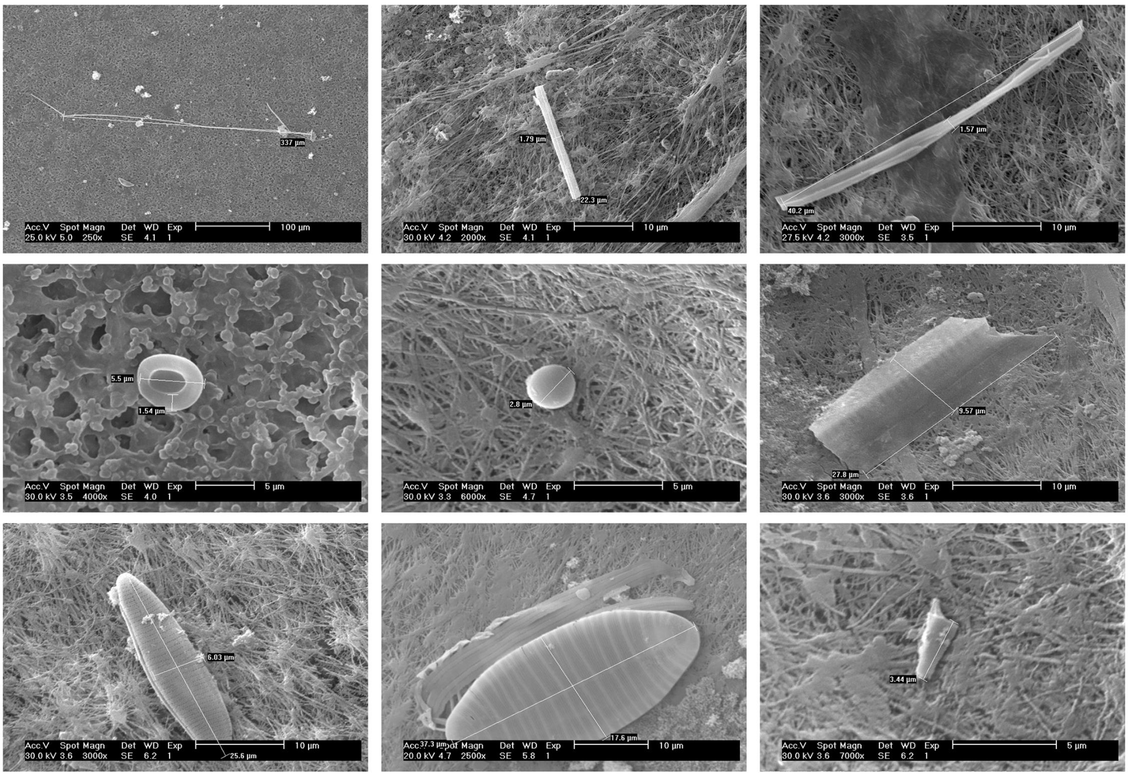

Figure 4. Microplastic images captured by SEM.

Table 1. Abundance and Measuring Methods of MPs and MPs Removal Efficiency in DWTPs.

STUDY	DIGESTION	DENSITY SEPARATION	FINEST SIEVE MESH OR FILTERS PORE SIZE (MM)	DETECTION	COUNTING	WATER SOURCE	DWTP STAGES	INFLUENT (MPS/L)	EFFLUENT (MPS/L)	REMOVAL EFFICIENCY
Pivokonsky et al. (2018)	-	-	0.2	micro-Raman micro-FTIR	SEM	Valley water reservoir	Coagulation/ flocculation, and sand filtration	1,473 ± 34	443 ± 10	70
						Water reservoir	Coagulation/ flocculation, sedimentation, sand filtration and GAC (Granular Activated Carbon) filtration	1,812 ± 35	338 ± 76	81
						River	Coagulation/ flocculation, flotation, sand filtration and GAC filtration	3,605 ± 497	628 ± 28	83
Wang et al. (2020)	WPO	-	0.2	micro-Raman	SEM	River	Coagulation/ flocculation, sedimentation, sand filtration, ozonation, and GAC filtration	6,614 ± 1,132	930 ± 71	82.1–88.6
Pivokonský et al. (2020)	-	-	0.2	micro-Raman	SEM	Dam	Coagulation/ flocculation, and sand filtration	23 ± 2	14 ± 1	40
						River	Coagulation/ flocculation, sedimentation, sand filtration, ozonation, and GAC filtration	1,266 ± 35	151 ± 4	88
Sarkar et al. (2021)	WPO	$ZnCl_2$	25	ATR FTIR	Optical microscope	River	Chlorination, coagulation, pulse clarification, and sand filtration	17.88	2.75	84.6
This study (2021)	WPO	-	0.45	micro-Raman	SEM	Dam	Coagulation/ flocculation, clarification, and sand filtration	1597.7 ± 270.3	260.5 ± 48.9	83.7

Table 2. MPs Removal Efficiency of Each Stage of DWTPs.

DWTP STAGES	MPS REMOVAL EFFICIENCY OF EACH STAGES (%)															
	BASED ON MPS SIZES				BASED ON MPS SHAPES				BASED ON MPS TYPES							TOTAL
	<10	10–50	50–100	100<	FIBER	FRAGMENT	OVAL	SPHERICAL	PE	PP	PVC	PET	PA	PS	NYLON 6	
Clarification	53.1	52.8	69.3	84.9	58.6	55.0	63.9	52.0	36.5	50.6	15.4	57.7	*	*	78.8	57.7
Filtration	55.5	63.9	80.2	86.1	52.7	67.1	76.2	68.3	74.3	72.5	61.5	76.9	*	61.5	*	61.5
Total efficiency	79.1	83.0	93.9	97.9	80.4	85.2	91.4	84.8	83.7	86.4	67.4	90.2	89.1	*	75.5	83.7

*The MPs removal efficiency based on MPs types at these stage cannot be measured because some type of MPs were not observed at all stages.

granular activated carbon (GAC) filtration remove MPs even more effectively. For example, in a DWTP that uses Coagulation/flocculation and sand filtration, only 40% of MPs were removed, while in another DWTP that uses Coagulation/flocculation, sedimentation, sand filtration, ozonation, and GAC filtration, the MPs removal efficiency was 88% (Pivokonský et al., 2020). Based on the results of (Pivokonsky et al., 2018; Pivokonský et al., 2020), it can be concluded that the DWTPs with direct filtration systems may have lower removal efficiencies than advanced DWTPs. In general, most DWTPs remove more than 80% of the influent's MPs.

Discharging rate of MPs into the water distribution system through DWTP

In this study, it was found that despite the effective removal of MPs in a DWTP and 85.8% removal efficiency, considering that this DWTP treats water with a capacity of 10,000 L/S, on average $2.25 \times 10^{11} \pm 4.23 \times 1010$ MPs are daily discharged into the water distribution system through the effluent of this DWTP.

Conclusion

MPs removal efficiency was investigated in this study. The MPs concentration in the influent, after clarifier, and effluent were 1597.7 ± 270.3, 676.2 ± 69.0, and 260.5 ± 48.9 MPs/L, respectively. Polypropylene, polyethylene, and polyethylene terephthalate were the most abundant polymers identified, respectively. The MPs removal efficiency in the clarification and filtration stage were 59.2% and 65.2%, respectively. The overall MPs removal efficiency of DWTP was 85.8%, which clarifier, and filtration stage removed 57.7%, and 26.0% of total MPs, respectively. The results of this study show that despite the effective removal of MPs in a DWTP, an average of $2.25 \times 10^{11} \pm 4.23 \times 1010$ MPs is discharged daily into the water distribution system through the effluent of this DWTP. This study contributes to the knowledge of MPs removal in DWTPs based on their characteristics such as size, morphology, and composition. More research is needed in the future to better understand the mass balance of MPs and even nanoplastics in DWTPs.

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Author Contributions

The authors certify that we have participated sufficiently in the intellectual content, conception, and design of this work or the analysis and interpretation of the data, as well as the writing of the manuscript, to take public responsibility for it.

Data Availability Statement

The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

Declartion of Conflicting Interests

The author(s) declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

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Ethics Approval and Consent to Participate

The project study related to this article has been evaluated ethically at Isfahan University of Medical Sciences (IR.MUI.RESEARCH.REC.1400.273). Also, the authors certify that we agree to publication this article.

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Notes

1. Polybrominated diphenyl ethers.
2. Polycyclic aromatic hydrocarbon.
3. Polychlorinated biphenyl

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