Detection and Analysis of Microfibers and Microplastics in Wastewater from a Textile Company

microplastics

MDPI

Article

Detection and Analysis of Microfibers and Microplastics in Wastewater from a Textile Company

Sinem Hazal Akyildiz [1], Rossana Bellopede [2], Hande Sezgin [3], Ipek Yalcin-Enis [3], Bahattin Yalcin [4] and Silvia Fiore [2,*]

[1] Department of Textile Engineering, Faculty of Technology, Marmara University, Göztepe Campus, 34722 Istanbul, Turkey
[2] Department of Engineering for Environment, Land and Infrastructures, Politecnico di Torino, 10129 Torino, Italy
[3] Department of Textile Engineering, Technologies and Design Faculty, Istanbul Technical University, Gumussuyu Campus, 34437 Istanbul, Turkey
[4] Department of Chemistry, Faculty of Science, Marmara University, Göztepe Campus, 34722 Istanbul, Turkey
* Correspondence: silvia.fiore@polito.it

Abstract: Textile wastewater is polluted by inorganic/organic substances, polymers, dyes, and microfibers (MFs), which are microplastics (MPs) and natural fibers. This work is aimed at the preliminary investigation of MFs and MPs in textile industrial wastewater, and at evaluating the removal efficiency of an on-site wastewater treatment plant (WWTP). Ten samples of inflows and outflows of the WWTP of a textile company (applying a physic-chemical process) have been analyzed. Firstly, the samples underwent a pretreatment with 15% hydrogen peroxide at 25 °C for 5 days to remove organic compounds. Secondly, the MFs were recovered from the aqueous phase by pre-screening centrifugation, density separation, and filtration as alternative options. Filtration obtained the best performances, compared to the other recovery processes. Thirdly, the MFs were counted through optical microscopy and the MPs were identified through micro-FTIR. The MFs amount in the inflow samples was in the range of 893–4452 MFs/L. The outflow samples (310–2404 MFs/L) exhibited a 38–65% reduction compared to the inflows, demonstrating that up to 62% of residual MFs can enter the sewer network or the receiving water body. Cotton and wool, and numerous MPs (acrylic, polyester, polypropylene, polyamide, and viscose/rayon) were identified in the inflow and outflow samples (with the only exception of "dense" viscose (rayon), not detected in the outflows, and probably retained by the WWTP with the sludge). This study, even if just preliminary, offers interesting hints for future research on MFs/MPs detection in textile wastewater, and on the performance of a full-scale WWT process for their removal.

Keywords: filtration; microfiber; microplastic; textile; wastewater

Citation: Akyildiz, S.H.; Bellopede, R.; Sezgin, H.; Yalcin-Enis, I.; Yalcin, B.; Fiore, S. Detection and Analysis of Microfibers and Microplastics in Wastewater from a Textile Company. *Microplastics* **2022**, *1*, 572–586. https://doi.org/10.3390/microplastics1040040

Academic Editor: Nicolas Kalogerakis

Received: 10 August 2022
Accepted: 28 September 2022
Published: 3 October 2022

1. Introduction

Microplastics (MPs) are particles smaller than 1 mm, according to ISO/TR 21960 [1], while the scientific literature classifies MPs as particles up to 5 mm [2–5]. MPs can have different shapes (fibers, granules, fragments, etc.) and they have been detected in aquatic ecosystems, urban runoff, and wastewater [6]. MPs can derive from many sources (e.g., tires wearing, clothes washing, etc.) [7,8]. MPs can enter the human body through food such as milk [9], honey [10], salt [11], seafood and fish [12], drinking water [13], and air [14,15], posing serious health risks [16]. The annual intake of MPs in the United States was estimated between 39,000 and 121,000 particles per person, based on the consumption of food, bottled water, and air inhalation [17]. MPs' capacity to interact with human body tissues may result in inflammatory lesions, and MPs' potential to influence immunological and metabolic illnesses has been hypothesized [18].

MPs' analysis in water typically involves three consequent steps [19], not yet standardized, as follows. Firstly, a pretreatment is used based on the use of reagents (e.g., hydrogen peroxide, Fenton's reagent, hydrochloric acid, sodium/potassium hydroxide, enzymes, etc.), and aimed at removing organic compounds [20]. Secondly, MPs are recovered from the aqueous phase to be analyzed [21,22], via density separation [23], vacuum filtration [24], or centrifugation [25]. Thirdly, MPs are quantified (usually through optical and stereo microscopy) and characterized through microscopy coupled with Fourier Transform Infrared (micro-FTIR), Near Infrared (NIR), Raman, Nuclear Magnetic Resonance (NMR) spectroscopy, Scanning Electron Microscopy (SEM) and Energy Dispersive X-ray Spectroscopy (EDS). Fourier-transform infrared (FTIR) spectroscopy is the most widely used method for detecting MPs, thanks to its repeatability and nondestructive nature [26]. Micro-FTIR can identify MPs smaller than 20 µm, which is a limitation of FTIR [27].

MPs are very common in the inflows of urban wastewater treatment plants (WWTPs) [28] and state-of-the-art WWT technologies are not able to produce MPs-free effluents [29–31]. MPs found in the inflows of urban WWTPs may be microfibers (MFs), granules, pellets, films, fragments, etc., and usually, the MPs detected in the effluents of urban WWTPs are MFs [28], as the other types tend to be eliminated in the settling phase [32]. An Italian study [33] showed that WWT can remove up to 84% of MPs and that every day 1.6 billion MPs are released into freshwater through the effluent, while the agricultural disposal of the sludge implied a daily deposition of 3.4 billion MPs into the soil. A study related to a Turkish WWTP [34] revealed 73–79% MPs' removal efficiency, based on a concentration of 1–6.5 million particles per day in the influent, and up to 1.5 million residual MPs per day in the effluent. A review [35] analyzing 15 studies reported an overall efficiency in MPs removal up to 90%. Another review analyzing the performances of 16 urban WWTPs all over the world [28] reported an overall efficiency in MPs' removal in the range of 60–99.9% depending on the applied technology. The highest MPs removal efficiency (99.9%) was reported for membrane bioreactor (MBR) processes [36,37]. A recent Italian study [38] analyzing three WWTPs demonstrated that MBR technology allows to greatly improve (up to 76%, compared to active sludge with or without primary settling) the concentration of MPs in the sludge. Considering the consequent phases of state-of-the-art WWT, mostly based on active sludge processes, it was reported [19,31] that 35–59% of MPs are removed through preliminary treatments, 50–98% via primary settling, up to 20% through secondary settling, and that about 2% residual MPs can be detected in the effluent. The high variability of the MPs' removal performances reported by literature is a key critical issue for the transferability of its results to the full scale. It should be mentioned that MPs-targeted WWT technologies are still in the research phase [19,28]. Standardization and/or harmonization of MPs' recovery and analysis procedures are urgently required to facilitate a critical analysis of the results of the available literature [19]. Moreover, inconsistencies have often been reported between the MPs concentrations adopted in the experimental studies with the ones found in the environment [39].

A relevant source of MPs in wastewater is textiles' washing [40] accounting for up to 35% of the total MPs identified in aquatic ecosystems [41] and the textile industry has a key role in introducing MPs and natural MFs (as wool, cotton, etc.) in the environment. A link between the concentration of MPs and MFs in freshwater and sediments in a specific local context and the intensity of textile industrial activities has been demonstrated [42–44]. Some studies describing Chinese textile industrial districts (holding the largest share of synthetic fibers' production in the world) [45] reported a concentration of MFs > 54,000/L in the wastewater [46] and a residual concentration in the effluent of 537 MFs/L; an average concentration of 362 MFs/L in the wastewater [47]; an average concentration of 334 MFs/L in the wastewater [30] and a residual concentration of 16 MFs/L in the effluent (95% removal). A concentration of 0.02 ± 0.02 g MPs/L in textile wastewater was reported by a Portuguese study referring to one single sample [48].

As mentioned before, extensive literature is available on MPs detection and analysis in urban wastewater and on the assessment of the efficiency of up-to-date WWT technologies in their removal. On the other hand, to our knowledge, the release of MPs in textile wastewater was previously explored by only a few studies [30,46–48], analyzing single samples collected from different facilities, and reporting highly variable data. In these references, the variability at the time of MPs concentration in textile wastewater was not considered, and the removal performances of the existing WWTPs have not been analyzed specifically referring to the applied technologies. The novelty of this study is based on several issues, as follows. Firstly, multiple samples collected at the inflow and outflow of a full-scale industrial WWTP located in a textile industry have been considered. This study was preliminary; the idea was to explore the variability of MPs' concentration in a specific facility for some months without yet performing a systematic sampling campaign. Secondly, this study allowed us to preliminary investigate the performances of a full-scale WWTP based on a coagulation/settling process, which is a technological solution commonly applied to the on-site treatment of industrial textile wastewater [49]. This study analyzed MPs through optical microscopy and micro-FTIR, considering 10 samples supplied by a Turkish textile company producing different types of synthetic fabrics. The company performs an on-site physic-chemical WWT process, and the samples have been collected for 5 months from its inflow and outflow. This study has two main aims: (i). investigating the amount and characteristics of MPs in the wastewater of a textile industry; (ii). assessing the performances of a full-scale physic-chemical WWT process towards MPs' removal. Moreover, this study pre-screened different options for the recovery of MPs from the pretreated aqueous samples, comparing centrifugation, density separation, and filtration.

2. Materials and Methods

2.1. Samples Origin and Pretreatment

A Turkish textile company producing wool, cotton, acrylic, polyamide, polyester, polypropylene, and viscose (rayon) fabrics, provided 10 wastewater samples (1 L) collected along 5 months (17 February, 23 March, and 8–13–14 June) in 2022. Before starting the sampling, all equipment was cleaned with distilled water and sterilized with ethanol. The bottles were manually filled with wastewater, and after being capped, they were covered with paraffin and stored in the refrigerator until the analysis to prevent bacterial growth. On these dates, 5 samples were collected at the inflow of the on-site WWTP, and 5 at the outflow. The inflow samples were made of the wastewater deriving from all the wet processes (dyeing, rinsing, softening, mechanical/chemical finishing) performed in the factory on the numerous types of fabrics produced. The outline of the on-site WWTP is based on a multi-phase physic-chemical process aimed at controlling pH, organic substances, and suspended solids through the addition of lime, iron-sulfate, and anionic polyelectrolyte in consequent stirred reactors, ending the process with a settling tank. The outflow samples were collected from the clarified effluent of the settling phase.

Before starting the processing, all equipment was cleaned with distilled water and sterilized with ethanol. Then the 1 L samples were pretreated for 5 days with 15% hydrogen peroxide (Merck KGaA, Darmstadt, Germany) at 25 °C [50–52].

2.2. MFs' Recovery

MFs' recovery from the pretreated aqueous phase was explored by testing centrifugation, density separation, and filtration [24,53] as alternative options. These tests had the mere purpose of pre-screening, and they were performed on three aliquots of one of the inflow samples (collected on March 23). All samples deriving from different recovery processes were vacuum filtered on 0.7 µm pore size glass fiber (GF) (Whatman, 47 mm diameter), and dried at 40 °C overnight before MFs/MPs' analysis. Centrifugation (6000 rpm for 20 min, Hermle LaborTechnik Z206A, Wehingen, Germany) generated two phases, i.e., sediments and supernatant, both filtered on GF. Density separation consisted of adding

150 g NaCl to the pretreated sample, and then, left to settle for a day; the resulting sink and float fractions were filtered on GF. Filtration was performed under vacuum on GF as above specified. The performances of the three different recovery processes have been evaluated through a visual count of the MFs on the GF filters (see Section 2.3).

2.3. MFs and MPs Analysis

The recovered MFs have been quantified by visual counting through a Leica ORTHOLUX II POL-MK (Wetzlar, Germany) optical microscope with normal and UV light, allowing the identification of fibers containing UV stabilizers. A Shimadzu AIM- 9000 Micro-FTIR was used to characterize the MFs, and MPs recovered on the GF filters in the wavelength range 700–4000 cm^{-1}. SHIMADZU–T-Polymer2 was the spectra library used.

3. Results and Discussion

3.1. Preliminary Comparison of the Different Recovery Processes

The result of the analyses of the GF filters resulting from the preliminarily tested recovery processes (Figure 1, 23 March sample) was as follows. MFs were surely collected by centrifugation, but some were still found in the supernatant. Density separation revealed that MFs were found in both sink and float fractions, due to the heterogeneous distribution of MFs' density. Finally, due to the high turbidity of the sample, an efficient filtration required multiple GF filters. In any case, filtration seemed the most effective recovery technique since it was able to collect MFs within a single phase, while the other techniques required two phases (more time-energy-consuming) and were not efficient. The achieved results are confirmed by literature, where different recovery processes have been applied to recover MPs from various types of samples. Specifically, density separation gave good results with sediments [51,53], centrifugation with sludge and bio-solids [54,55]. On the other hand, filtration achieved the best performance with aqueous samples [23,56,57]. In the study by Lares et al. [58], municipal wastewater and digested sludge samples underwent six different procedures (filtering, wet peroxide oxidation, degradation with KOH, oil extraction, density separation, and drying), previously used in MPs investigations in various media. The procedures were assessed by analyzing recovery rates, time consumption, benefits, and drawbacks. Filtration with size fractionation was found to be the most effective method for both wastewater and sludge samples, with 91.4% and 92.9% MPs recovery rates, respectively [58]. In a similar way, a review by Liu et al. [54] focused on current research on the presence and transmission of MPs in WWTPs and compared sampling and separation methods and analytical techniques for identifying and characterizing MPs. The review emphasized that filtration is the recommended method to remove and separate MPs from wastewater and sludge samples [59].

It is also interesting to observe that this research considered MFs and MPs larger than 0.7 μm, as all samples were filtered on 0.7 μm pore size GF (see Section 2.2). Literature reports that various materials (quartz, glass, nitrocellulose, cellulose, polycarbonate) and pore dimensions (from 0.2 to 12 μm) were used to recover MFs and MPs [51,53,55,56]. The filter composition and porosity were chosen depending on the analyzed sample (e.g., sludge, water, solid) and the analytical technique adopted for the identification. It should be noticed that, on one hand, the filter discards the MFs/MPs having dimensions below its porosity; on another hand, the accuracy of their identification is less reliable with small particles [60,61]. For these reasons, the Joint Research Centre of the European Commission [62] suggested a cut-off of particles smaller than 0.1 mm.

3.2. MPs Amount and Characterization

Depending on the turbidity of the samples, multiple GF filters (1 to 5) were necessary to recover MFs from the aqueous phase; thus, Table 1 shows variable numbers of filters (F) for each sample. The number of MFs was visually measured only on samples pretreated and vacuum filtered, i.e., the GF filters obtained from the tests on centrifugation and density separation were excluded from this measurement (see Section 2.2). This implies that the

count of MFs in the inflow sample collected on March 23 was performed only on one third of its volume; therefore, the sum of the MFs collected on the filters was multiplied by three to obtain the total estimated number of MFs for that sample.

Table 1. MFs quantification in the inflow and outflow samples pretreated and vacuum filtered.

Samples	17 February	23 March	8 June	13 June	14 June
Inflow	F1 = 447 F2 = 446 Total = 893	F1 = 268 F2 = 449 F3 = 767 Total * = 4452	F1 = 1921 F2 = 834 Total = 2755	F1 = 870 F2 = 539 Total = 1409	F1 = 1022 F2 = 642 Total = 1664
Outflow	F1 = 310	F1 = 491 F2 = 416 F3 = 428 F4 = 635 F5 = 434 Total = 2404	F1 = 625 F2 = 375 Total = 1000	F1 = 476 F2 = 391 Total = 867	F1 = 343 F2 = 377 Total = 720
% Decrease	65	46	63	38	56

* see Section 3.2.

The results of MFs visual count through optical microscopy (Table 1 and Figures 1 and 2) showed that the average amount of MFs in the analyzed samples was for the inflow samples in the range 893–4452 MFs/L, while for the outflow samples in the range 310–2404 MFs/L. It should be noticed that the number of MFs in the inflow and outflow of the on-site WWTP is highly variable in the analyzed samples. These values are higher compared to the few data reported by literature (circa 300 MFs/MPs per L) [30,47].

Considering the MFs' removal efficiency, the obtained values were again highly variable and between 38 and 65% (average 54 $\pm$ 11%), with the average and three values out of five (Table 1) in agreement with the range reported by literature for primary settling (50–98%) [19,31].

For characterization, the MFs detected through an optical microscope were measured one by one, obtaining fiber lengths ranging from 10 µm to 5 mm in all samples (Figure 3).

A high variability in the distribution of dimensions of MFs can be observed among the different samples. While in the samples of 17 February, 23 March, and 8 June the dimensions (both for inflow and outflow) of MFs are mainly in the range between 10 µm and 1 mm, in the samples of 13 and 14 June the distribution of dimensions in inflow samples is more homogenous, reaching a percentage of 20% of MFs of dimension between 1–2 mm (13 and 14 June samples). However, in all the samples the MFs with dimensions minor than 1 mm are predominant both in inflow and in outflow. The highest amount of MFs in the range between 10 µm and 0.1 mm has been detected in the inflow sample of 17 February (25% in comparison with 8–13% in the other samples) and the number increases in the outflow sample collected in the same data (reaching 30%). The MFs length class between 0.1 mm and 0.5 mm is for the samples 17 February, 23 March, and 8 June most abundant, reaching the maximum of 38% in outflow (17 February, 8 June) and 36% in inflow (17 February, 8 June). The 13 June sample is characterized by the most abundant MFs length class respectively of 0.5–1 mm for inflow (22%) and of 0.1–0.5 mm for outflow (32%). The 14 June sample is characterized by a maximum of length MFs in the length class 2–3 mm for the inflow sample (20.0%) and 10 µm-0.1 mm for the outflow sample (33%).

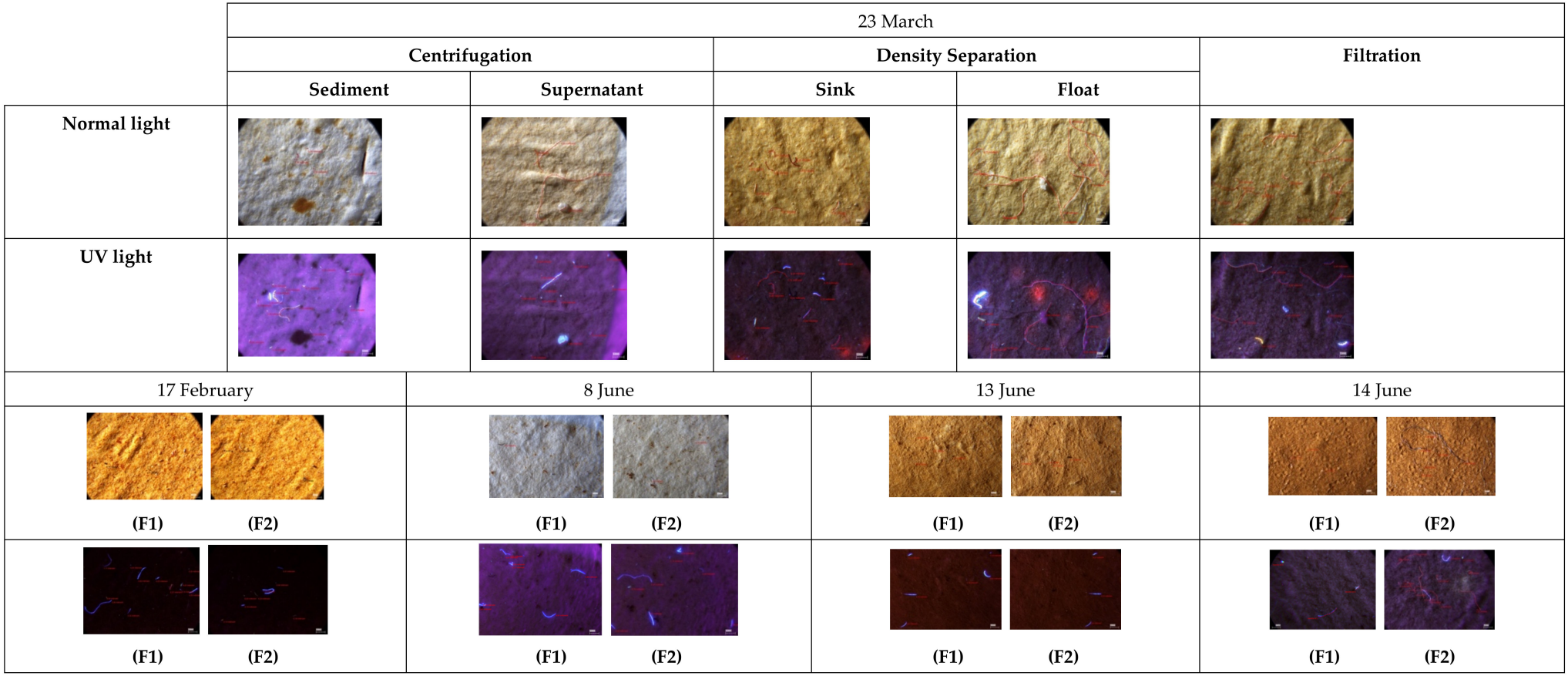

Figure 1. Microscopic images (normal and UV light) of the filters deriving from the inflow samples.

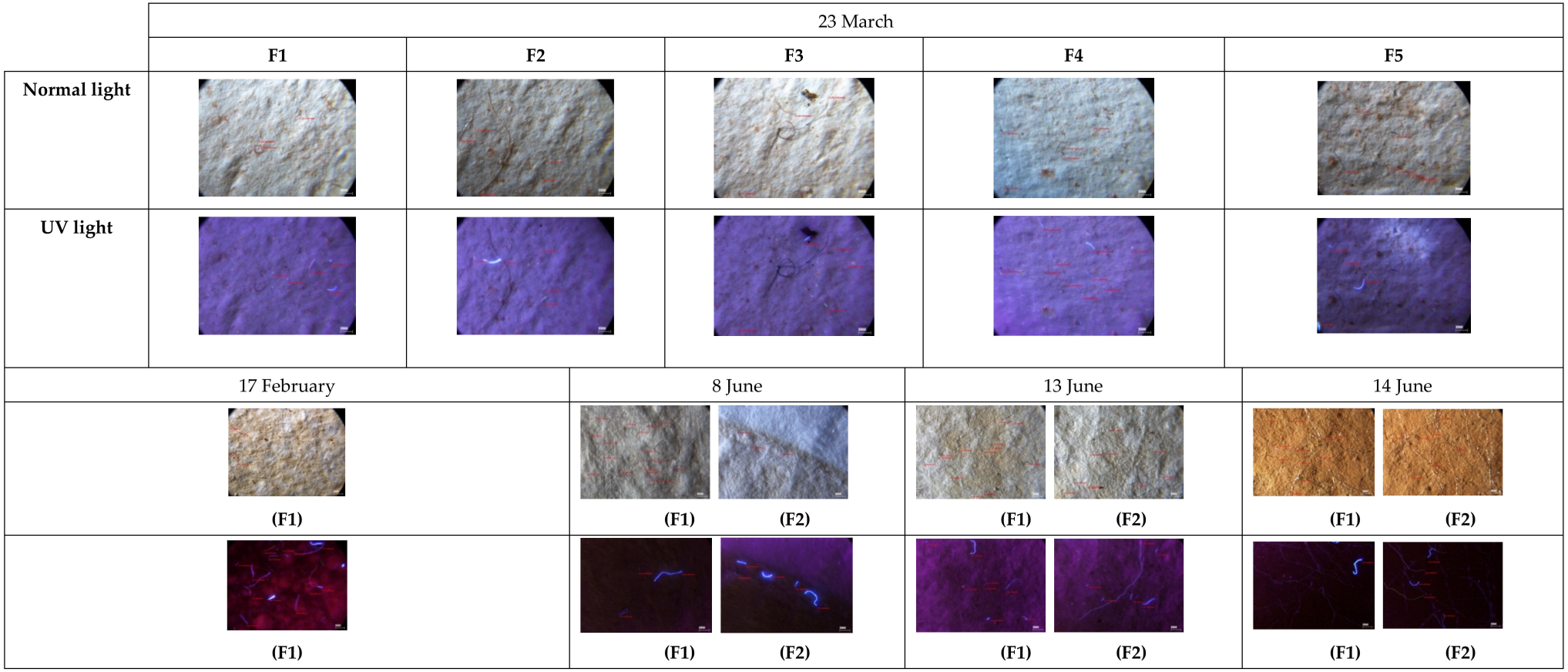

Figure 2. Microscopic images (normal and UV light) of the filters deriving from the outflow samples.

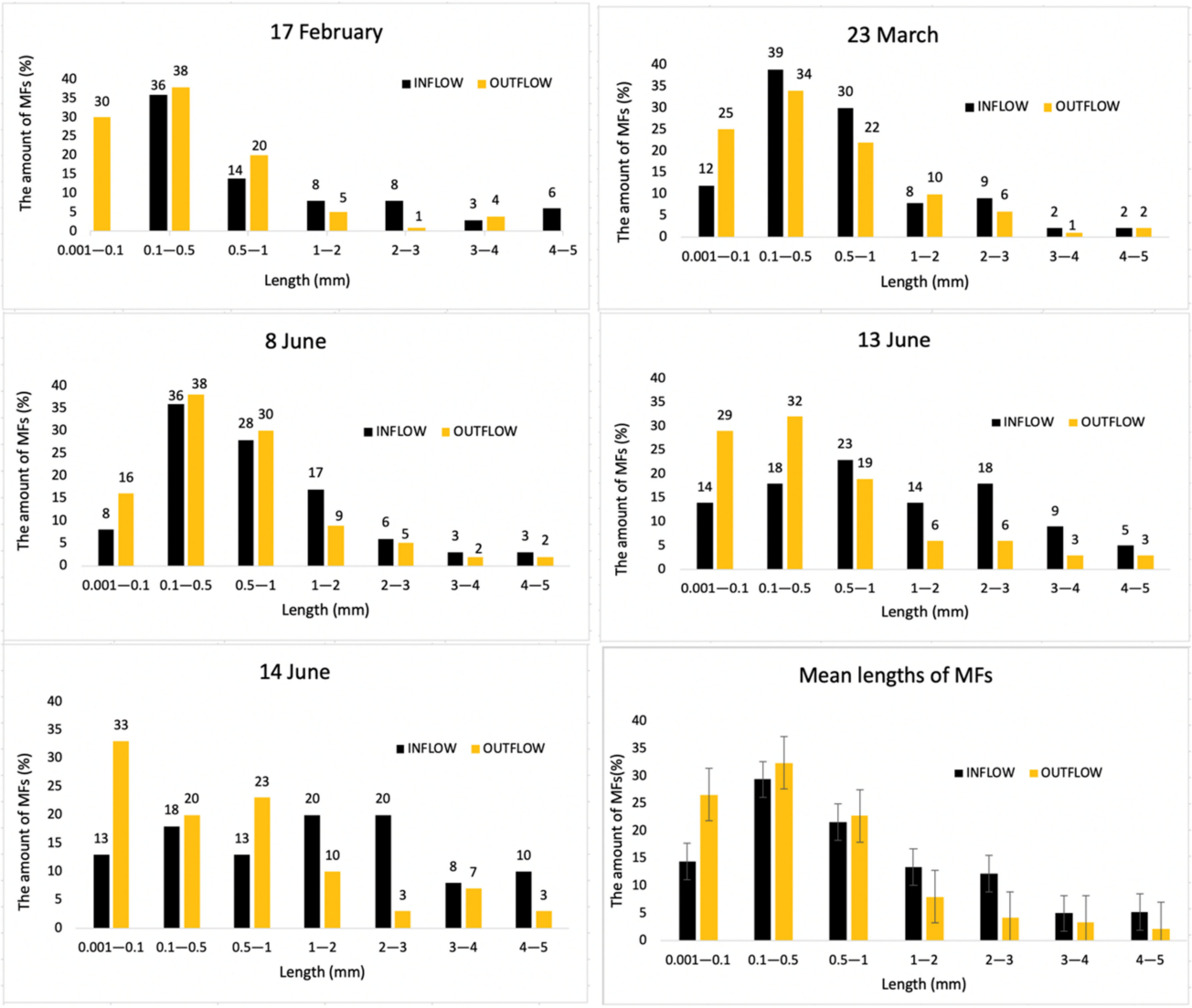

Figure 3. Distribution of MFs' lengths in the inflow and outflow samples.

Comparing the length of the MFs in the inflow and outflow samples for each sampling date, overall the outflow samples exhibited a higher amount (82.0 ± 6.1%) of particles having dimensions below 1 mm compared to the inflow samples (64.8 ± 8.3%), confirming that larger particles tend to settle with the chemical sludge in the on-site WWTP.

The results of micro-FTIR analysis (Table 2) allowed us to identify the recovered MFs in the inflow samples as cotton and wool, and numerous MPs were made of acrylic, polyester, polypropylene, polyamide, and viscose (rayon) (Figure 4). On the other hand, the MFs detected in the outflow samples were cotton and wool, and fewer MPs made of acrylic, polyester, polypropylene, and polyamide, with only viscose (rayon) not detected. These results are due to the different densities of the textile fibers. When considering natural fibers, the density of cotton fiber ranges from 1.50–1.54 g/cm^3; whereas, that of wool fiber is 1.30 g/cm^3. The densities/ranges for acrylic, polyamide, polyester, and polypropylene fibers are 1.52, 1.02–1.18, 1.22–1.38, and 0.95–0.96 g/cm^3, respectively [36]. Certain regenerated fibers, such as viscose (rayon), can have a significantly larger density range (1.53–4.50 g/cm^3) [63]. These are quite dense compared to other fibers and were not detected in the outflow samples probably because they settled with the sludge in the textile company's WWTP.

Table 2. Types of MFs and MPs identified in the considered samples.

Samples	17 February	23 March	8 June	13 June	14 June
Inflow	Acrylic Cotton Polyester	Acrylic Cotton Polyester Polypropylene Viscose (rayon) Wool	Acrylic Cotton Polyamide Polyester Viscose (rayon)	Acrylic Cotton Polyester Viscose (rayon)	Acrylic Cotton Polyester Viscose (rayon)
Outflow	Acrylic Cotton	Cotton Polyester Polypropylene Wool	Cotton Polyamide Polyester	Acrylic Cotton Polyester	Cotton Polyester

In detail, the following fibers were detected through micro-FTIR (Figure 4): acrylic (C–H stretching at 2924–2853, C=O stretching at 1734, CΞN stretching at 2242, and C–C stretch in-ring at 1452); cotton (O-H stretching at 3300, C–H stretching at 2896, C=O stretching at 1730, C–H bending at 1428, C–C, C–O, and C–O–C stretching at 1030); polyamide (N–H stretching at 3293, C–H stretching at 2932–2857, C=O stretching at 1631, C–H stretching at 1536–1460, and C–N stretching at 1373); polyester (C–H stretching at 2968–2908, C=O stretching at 1710, aromatic ring at 1504–1405, the carboxylic acid (C–O) at 1242, ester (O=C–O–C) at 1093, and aromatic (C–H) at 718); polypropylene (C-H stretching at 2952–2918–2823, C–H bending at 1455–1375, C–C bending at 1165); viscose(rayon) (O–H stretching at 3450, C–H stretching at 2900, N–H bending at 1655–1545, and C–O stretching at 1050); and wool (O–H stretching at 3300 and C–N stretching at 1227–1350–1506).

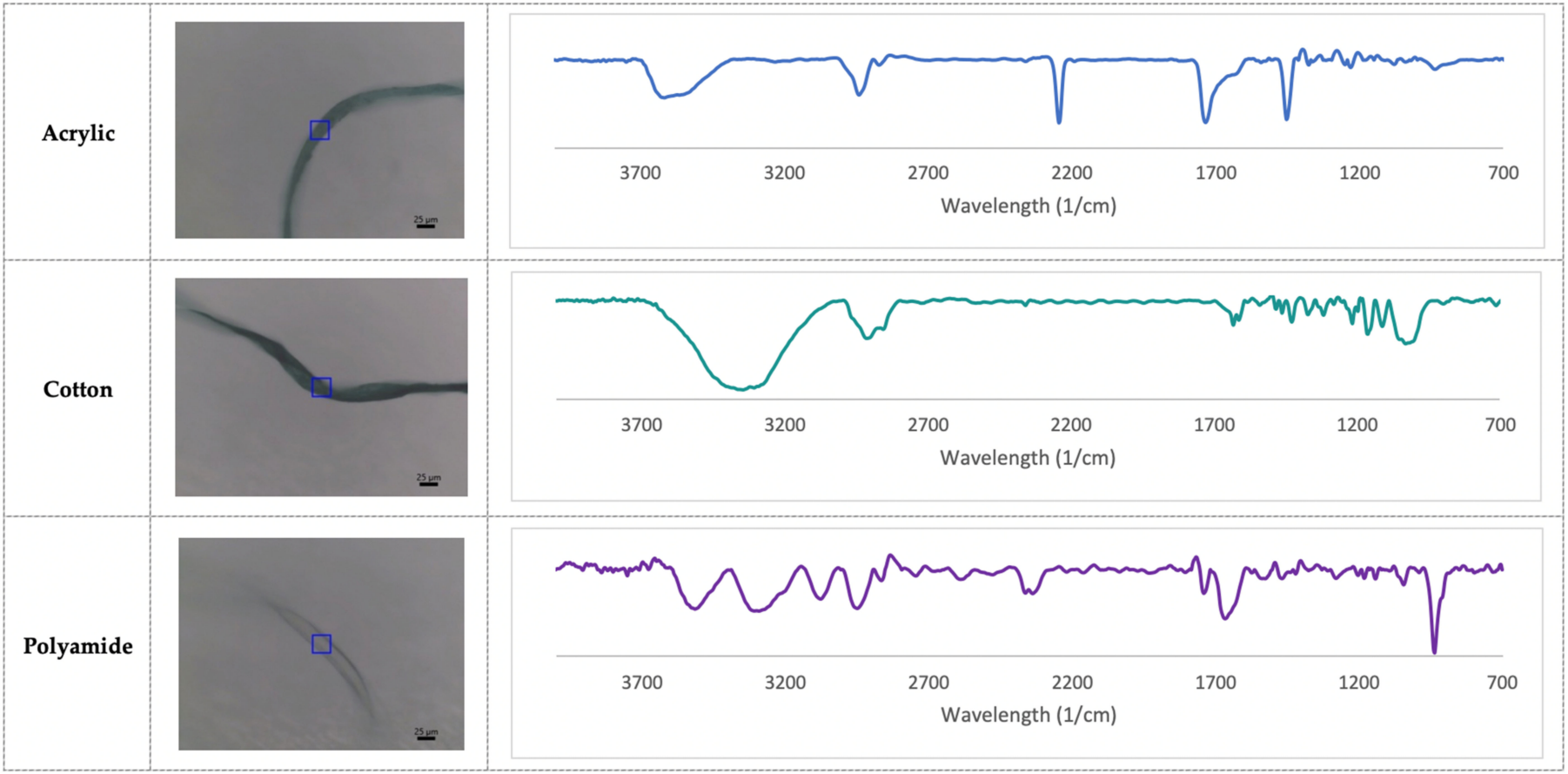

Figure 4. *Cont.*

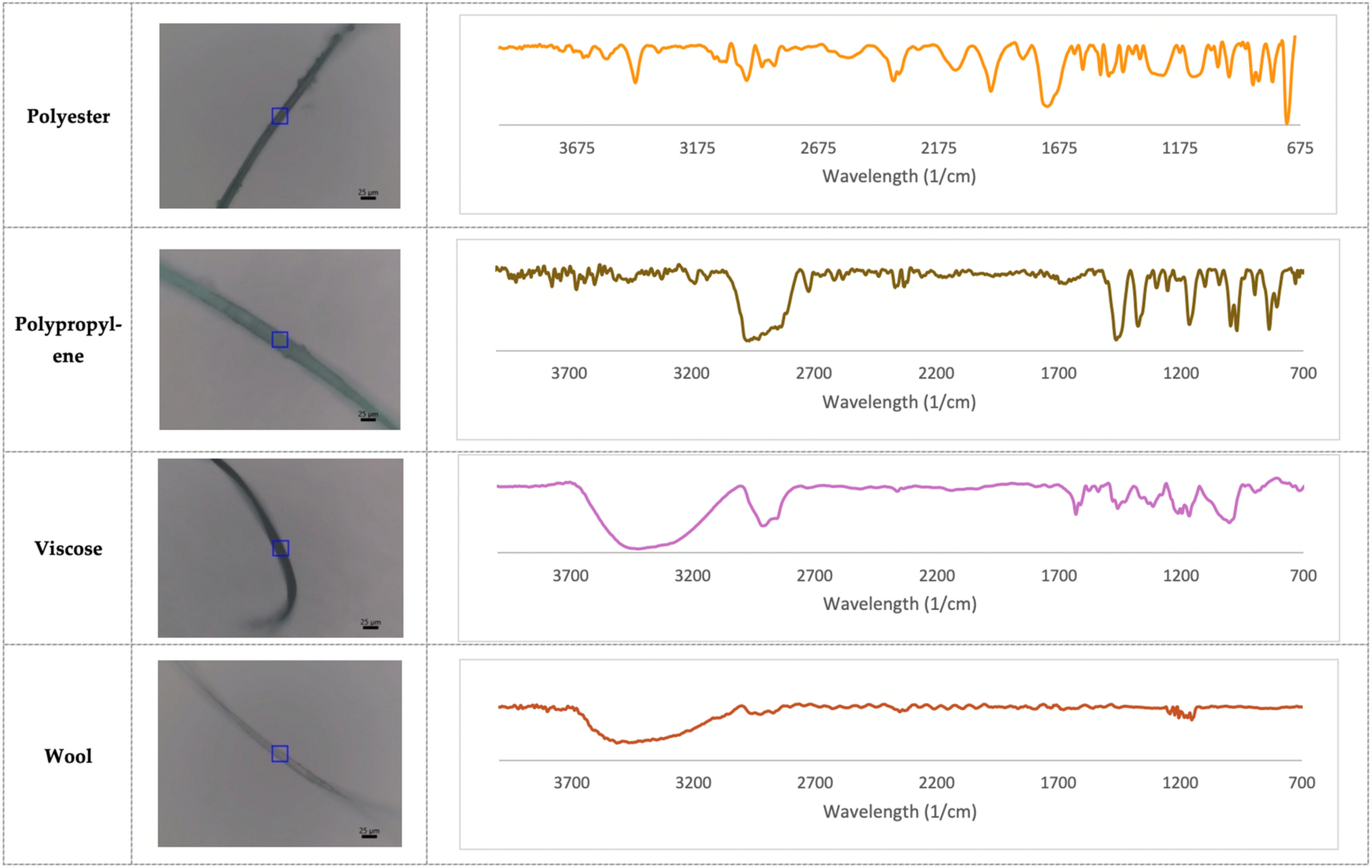

Figure 4. Results of the Micro-FTIR analyses on the recovered MFs and MPs.

4. Conclusions

This study had two objectives: preliminarily investigating MFs/MPs amount in wastewater samples collected at a Turkish textile company producing natural and synthetic fabrics; and evaluating the removal efficiency of an on-site WWTP performing a physic-chemical process based on multi-coagulation and settling. A total of 10 samples, 5 from the inflow and 5 from the outflow of the WWTP, have been collected over 6 months. The samples were pretreated to eliminate organic compounds and the MFs were recovered through vacuum filtration. With regards to the first objective, the MFs amount in the inflow samples was in the range of 893–4452 MFs/L, exhibiting a wide variability within the analyzed samples. Considering the second objective, the outflow samples demonstrated a 38–65% reduction in MFs compared to the inflows, in line with the performances reported by literature for primary settling applied in full-scale WWTPs. With regards to the MFs' length, the outflow samples exhibited a higher amount (82.0 $\pm$ 6.1%) of particles having dimensions below 1 mm compared to the inflow samples (64.8 $\pm$ 8.3%), confirming that larger particles tend to settle with the sludge. Cotton and wool and numerous synthetic fibers (acrylic, polyester, polypropylene, polyamide, and viscose (rayon)) were identified in the inflow and outflow samples. The only exception was that viscose (rayon) was not detected in the outflows and was probably retained by the WWTP with the sludge because of its higher density compared to other fibers.

These results demonstrated that, within the considered samples, the on-site WWT facility exhibited unsatisfactory performances of about MPs removal. Up to 62% o residual MFs can enter the sewer network, and thus, be a burden for the local urban WWTP. In the worst case, if a direct discharge in the surface water is foreseen, the residual MFs/MPs can pollute the receiving water body. The textile company providing the samples could improve the performance of the on-site WWT about MFs/MPs removal by enhancing the coagulation/settling process and increasing the coagulants' dose, though a larger amount of chemical sludge to be disposed of should be accounted for. This study has the characteristics of a preliminary investigation, considering the limited number of samples analyzed. However, it may provide some interesting hints for future research on the topic.

Author Contributions: Conceptualization, S.F. and R.B.; methodology, S.F. and R.B.; validation, S.H.A.; formal analysis, S.H.A.; investigation, S.H.A. and R.B.; resources, S.H.A. and R.B.; data curation, S.H.A.; writing—original draft preparation, S.H.A.; writing—review and editing, all authors; visualization, S.H.A.; supervision, S.F., R.B. and H.S.; project administration, B.Y. and I.Y.-E.; funding acquisition, H.S. All authors have read and agreed to the published version of the manuscript.

Funding: The research was supported through internal resources.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Acknowledgments: The authors gratefully acknowledge ERASMUS+ support for the mobility of Akyildiz, S.H. to Politecnico di Torino, Italy for 6 months (February–July 2022).

Conflicts of Interest: The authors declare no conflict of interest.

References

1. *ISO TR 21960 Plastics*; Environmental Aspects. State of Knowledge and Methodologies. ISO Plastics: Mount Vernon, NY, USA, 2020.
2. Crawford, C.B.; Quinn, B. *Microplastic Pollutants*; Elsevier Limited: Amsterdam, The Netherlands, 2016.
3. De Lucia, G.A.; Vianello, A.; Camedda, A.; Vani, D.; Tomassetti, P.; Coppa, S.; Palazzo, L.; Amici, M.; Romanelli, G.; Zampetti, G.; et al. Sea Water Contamination in the Vicinity of the Italian Minor Islands Caused by Microplastic Pollution. *Water* **2018**, *10*, 1108. [CrossRef]
4. Liu, D.; Zheng, Y.; Chen, L.; Wen, D. Prevalence of small-sized microplastics in coastal sediments detected by multipoint confocal micro-Raman spectrum scanning. *Sci. Total Environ.* **2022**, *831*, 154741. [CrossRef] [PubMed]

5. Khan, L.; Ghias, S.; Zafar, M.I.; Alhodaib, A.; Fatima, H.; Ur-Rehman, T.; Waseem, A.; Howari, H. Exploration of microplastic pollution with particular focus on source identification and spatial patterns in riverine water, sediment and fish of the Swat River, Pakistan. *RSC Adv.* **2022**, *12*, 9556–9566. [CrossRef] [PubMed]
6. Ziajahromi, S.; Neale, P.A.; Rintoul, L.; Leusch, F.D.L. Wastewater treatment plants as a pathway for microplastics: Development of a new approach to sample wastewater-based microplastics. *Water Res.* **2017**, *112*, 93–99. [CrossRef] [PubMed]
7. Wang, T.; Li, B.; Yu, W.; Zou, X. Microplastic pollution and quantitative source apportionment in the Jiangsu coastal area, China. *Mar. Pollut. Bull.* **2021**, *166*, 112237. [CrossRef]
8. Luo, Z.; Zhou, X.; Su, Y.; Wang, H.; Yu, R.; Zhou, S.; Xu, E.G.; Xing, B. Environmental occurrence, fate, impact, and potential solution of tire microplastics: Similarities and differences with tire wear particles. *Sci. Total Environ.* **2021**, *795*, 148902. [CrossRef]
9. Kutralam-Muniasamy, G.; Pérez-Guevara, F.; Elizalde-Martínez, I.; Shruti, V.C. Branded Milks—Are They Immune from Microplastics Contamination? *Sci. Total Environ.* **2020**, *714*, 136823. [CrossRef]
10. Liebezeit, G.; Liebezeit, E. Non-pollen particulates in honey and sugar. *Food Addit. Contam. Part A* **2013**, *30*, 2136–2140. [CrossRef]
11. Yang, D.; Shi, H.; Li, L.; Li, J.; Jabeen, K.; Kolandhasamy, P. Microplastic Pollution in Table Salts from China. *Environ. Sci. Technol.* **2015**, *49*, 13622–13627. [CrossRef]
12. Smith, M.; Love, D.C.; Rochman, C.M.; Neff, R.A. Microplastics in Seafood and the Implications for Human Health. *Curr. Environ. Health Rep.* **2018**, *5*, 375–386. [CrossRef]
13. Koelmans, A.A.; Nor, N.H.M.; Hermsen, E.; Kooi, M.; Mintenig, S.M.; De France, J. Microplastics in freshwaters and drinking water: Critical review and assessment of data quality. *Water Res.* **2019**, *155*, 410–422. [CrossRef] [PubMed]
14. Catarino, A.I.; Macchia, V.; Sanderson, W.G.; Thompson, R.C.; Henry, T.B. Low levels of microplastics (MP) in wild mussels indicate that MP ingestion by humans is minimal compared to exposure via household fibres fallout during a meal. *Environ. Pollut.* **2018**, *237*, 675–684. [CrossRef] [PubMed]
15. Barboza, L.G.A.; Vethaak, A.D.; Lavorante, B.R.; Lundebye, A.-K.; Guilhermino, L. Marine microplastic debris: An emerging issue for food security, food safety and human health. *Mar. Pollut. Bull.* **2018**, *133*, 336–348. [CrossRef] [PubMed]
16. Vethaak, A.D.; Legler, J. Microplastics and human health. *Science* **2021**, *371*, 672–674. [CrossRef]
17. Cox, K.D.; Covernton, G.A.; Davies, H.L.; Dower, J.F.; Juanes, F.; Dudas, S.E. Human Consumption of Microplastics. *Environ. Sci. Technol.* **2019**, *53*, 7068–7074. [CrossRef]
18. Prata, J.C.; da Costa, J.P.; Lopes, I.; Duarte, A.C.; Rocha-Santos, T. Environmental exposure to microplastics: An overview on possible human health effects. *Sci. Total Environ.* **2019**, *702*, 134455. [CrossRef]
19. Sun, J.; Dai, X.; Wang, Q.; van Loosdrecht, M.C.; Ni, B.-J. Microplastics in wastewater treatment plants: Detection, occurrence and removal. *Water Res.* **2019**, *152*, 21–37. [CrossRef]
20. Cole, M.; Webb, H.; Lindeque, P.K.; Fileman, E.S.; Halsband, C.; Galloway, T.S. Isolation of microplastics in biota-rich seawater samples and marine organisms. *Sci. Rep.* **2014**, *4*, 4528. [CrossRef]
21. Liu, M.; Lu, S.; Chen, Y.; Cao, C.; Bigalke, M.; He, D. Analytical Methods for Microplastics in Environments: Current Advances and Challenges. In *Microplastics in Terrestrial Environments*; Springer: Berlin/Heidelberg, Germany, 2020; pp. 3–24. [CrossRef]
22. Nguyen, B.; Claveau-Mallet, D.; Hernandez, L.M.; Xu, E.G.; Farner, J.M.; Tufenkji, N. Separation and Analysis of Microplastics and Nanoplastics in Complex Environmental Samples. *Accounts Chem. Res.* **2019**, *52*, 858–866. [CrossRef]
23. Fok, L.; Lam, T.W.L.; Li, H.-X.; Xu, X.-R. A meta-analysis of methodologies adopted by microplastic studies in China. *Sci. Total Environ.* **2019**, *718*, 135371. [CrossRef]
24. Nabi, I.; Bacha, A.-U.; Zhang, L. A review on microplastics separation techniques from environmental media. *J. Clean. Prod.* **2022**, *337*, 130458. [CrossRef]
25. Elkhatib, D.; Oyanedel-Craver, V. A Critical Review of Extraction and Identification Methods of Microplastics in Wastewater and Drinking Water. *Environ. Sci. Technol.* **2020**, *54*, 7037–7049. [CrossRef] [PubMed]
26. Tirkey, A.; Upadhyay, L.S.B. Microplastics: An overview on separation, identification and characterization of microplastics. *Mar. Pollut. Bull.* **2021**, *170*, 112604. [CrossRef] [PubMed]
27. Löder, M.G.J.; Gerdts, G. Methodology Used for the Detection and Identification of Microplastics—A Critical Appraisal. In *Marine Anthropogenic Litter*; Springer: Berlin/Heidelberg, Germany, 2015; pp. 201–227. [CrossRef]
28. Ngo, P.L.; Pramanik, B.K.; Shah, K.; Roychand, R. Pathway, classification and removal efficiency of microplastics in wastewater treatment plants. *Environ. Pollut.* **2019**, *255*, 113326. [CrossRef]
29. Li, J.; Liu, H.; Chen, J.P. Microplastics in freshwater systems: A review on occurrence, environmental effects, and methods for microplastics detection. *Water Res.* **2018**, *137*, 362–374. [CrossRef]
30. Xu, X.; Hou, Q.; Xue, Y.; Jian, Y.; Wang, L. Pollution characteristics and fate of microfibers in the wastewater from textile dyeing wastewater treatment plant. *Water Sci. Technol.* **2018**, *78*, 2046–2054. [CrossRef]
31. Gies, E.A.; LeNoble, J.L.; Noël, M.; Etemadifar, A.; Bishay, F.; Hall, E.R.; Ross, P.S. Retention of microplastics in a major secondary wastewater treatment plant in Vancouver, Canada. *Mar. Pollut. Bull.* **2018**, *133*, 553–561. [CrossRef]
32. Castelluccio, S.; Alvim, C.B.; Bes-Piá, M.A.; Mendoza-Roca, J.A.; Fiore, S. Assessment of Microplastics Distribution in a Biological Wastewater Treatment. *Microplastics* **2022**, *1*, 141–155. [CrossRef]
33. Magni, S.; Binelli, A.; Pittura, L.; Giacomo, C.; Della, C.; Carla, C.; Regoli, F. The fate of microplastics in an Italian Wastewater Treatment Plant. *Sci. Total Environ.* **2019**, *652*, 602–610. [CrossRef]

34. Gündoğdu, S.; Çevik, C.; Güzel, E.; Kilercioğlu, S. Microplastics in municipal wastewater treatment plants in Turkey: A comparison of the influent and secondary effluent concentrations. *Environ. Monit. Assess.* **2018**, *190*, 626. [CrossRef]
35. Cristaldi, A.; Fiore, M.; Zuccarello, P.; Conti, G.O.; Grasso, A.; Nicolosi, I.; Copat, C.; Ferrante, M. Efficiency of Wastewater Treatment Plants (WWTPs) for Microplastic Removal: A Systematic Review. *Int. J. Environ. Res. Public Health* **2020**, *17*, 8014. [CrossRef] [PubMed]
36. Talvitie, J.; Mikola, A.; Koistinen, A.; Setälä, O. Solutions to microplastic pollution—Removal of microplastics from wastewater effluent with advanced wastewater treatment technologies. *Water Res.* **2017**, *123*, 401–407. [CrossRef] [PubMed]
37. Zeri, C.; Adamopoulou, A.; Koi, A.; Koutsikos, N.; Lytras, E.; Dimitriou, E. Rivers and Wastewater-Treatment Plants as Microplastic Pathways to Eastern Mediterranean Waters: First Records for the Aegean Sea, Greece. *Sustainability* **2021**, *13*, 5328. [CrossRef]
38. Di Bella, G.; Corsino, S.F.; De Marines, F.; Lopresti, F.; La Carrubba, V.; Torregrossa, M.; Viviani, G. Occurrence of Microplastics in Waste Sludge of Wastewater Treatment Plants: Comparison between Membrane Bioreactor (MBR) and Conventional Activated Sludge (CAS) Technologies. *Membranes* **2022**, *12*, 371. [CrossRef]
39. Weis, J.S.; Palmquist, K.H. Reality Check: Experimental Studies on Microplastics Lack Realism. *Appl. Sci.* **2021**, *11*, 8529. [CrossRef]
40. De Falco, F.; Gullo, M.P.; Gentile, G.; Di Pace, E.; Cocca, M.; Gelabert, L.; Brouta-Agnésa, M.; Rovira, A.; Escudero, R.; Villalba, R.; et al. Evaluation of microplastic release caused by textile washing processes of synthetic fabrics. *Environ. Pollut.* **2018**, *236*, 916–925. [CrossRef]
41. Xu, C.; Zhang, B.; Gu, C.; Shen, C.; Yin, S.; Aamir, M.; Li, F. Are We Underestimating the Sources of Microplastic Pollution in Terrestrial Environment? *J. Hazard. Mater.* **2020**, *400*, 123228. [CrossRef] [PubMed]
42. Deng, H.; Wei, R.; Luo, W.; Hu, L.; Li, B.; Di, Y.; Shi, H. Microplastic pollution in water and sediment in a textile industrial area. *Environ. Pollut.* **2020**, *258*, 113658. [CrossRef]
43. Xu, C.; Zhou, G.; Lu, J.; Shen, C.; Dong, Z.; Yin, S.; Li, F. Spatio-vertical distribution of riverine microplastics: Impact of the textile industry. *Environ. Res.* **2022**, *211*, 112789. [CrossRef]
44. Cesa, F.S.; Turra, A.; Baruque-Ramos, J. Synthetic fibers as microplastics in the marine environment: A review from textile perspective with a focus on domestic washings. *Sci. Total Environ.* **2017**, *598*, 1116–1129. [CrossRef]
45. Henry, B.; Laitala, K.; Klepp, I.G. *Microplastic Pollution from Textiles: A Literature Review*; Consumption Research Norway: Oslo, Norway, 2018. Available online: https://oda.oslomet.no/oda-xmlui/handle/20.500.12199/5360 (accessed on 25 July 2022).
46. Zhou, H.; Zhou, L.; Ma, K. Microfiber from textile dyeing and printing wastewater of a typical industrial park in China: Occurrence, removal and release. *Sci. Total Environ.* **2020**, *739*, 140329. [CrossRef] [PubMed]
47. Chan, C.K.; Park, C.; Chan, K.M.; Mak, D.C.; Fang, J.K.; Mitrano, D.M. Microplastic Fibre Releases from Industrial Wastewater Effluent: A Textile Wet-Processing Mill in China. *Environ. Chem.* **2021**, *18*, 93–100. [CrossRef]
48. Magalhães, S.; Alves, L.; Romano, A.; Medronho, B.; Rasteiro, M.D.G. Extraction and Characterization of Microplastics from Portuguese Industrial Effluents. *Polymers* **2022**, *14*, 2902. [CrossRef] [PubMed]
49. Tchobanoglous, G.; Stensel, H.D.; Tsuchihashi, R.; Burton, G.; Abu-Orf, M.; Bowden, H.; von Pfuel, S. *Wastewater Engineering: Treatment and Resource Recovery*; McGraw-Hill Education: New York, NY, USA, 2014.
50. Akyildiz, S.H.; Bellopede, R.; Fiore, S.; Yalcin, B.; Sezgin, H.; Yalcin-Enis, İ. Preliminary Assessment of Microfibers Amount in Textile Wastewater. In Proceedings of the 6th Symposium on Circular Economy and Urban Mining, Capri, Italy, 18–20 May 2022; CISA Publisher: Padua, Italy, 2022.
51. Zhang, C.; Zhou, H.; Cui, Y.; Wang, C.; Li, Y.; Zhang, D. Microplastics in offshore sediment in the Yellow Sea and East China Sea, China. *Environ. Pollut.* **2018**, *244*, 827–833. [CrossRef] [PubMed]
52. Mathalon, A.; Hill, P. Microplastic fibers in the intertidal ecosystem surrounding Halifax Harbor, Nova Scotia. *Mar. Pollut. Bull.* **2014**, *81*, 69–79. [CrossRef]
53. Balestra, V.; Bellopede, R. Microplastic pollution in show cave sediments: First evidence and detection technique. *Environ. Pollut.* **2021**, *292*, 118261. [CrossRef]
54. Monteiro, S.S.; Rocha-Santos, T.; Prata, J.C.; Duarte, A.C.; Girão, A.V.; Lopes, P.; Cristovão, T.; da Costa, J.P. A Straightforward Method for Microplastic Extraction from Organic-Rich Freshwater Samples. *Sci. Total Environ.* **2022**, *815*, 152941. [CrossRef]
55. Zhou, Y.; Liu, X.; Wang, J. Characterization of microplastics and the association of heavy metals with microplastics in suburban soil of central China. *Sci. Total Environ.* **2019**, *694*, 133798. [CrossRef]
56. Raju, S.; Carbery, M.; Kuttykattil, A.; Senthirajah, K.; Lundmark, A.; Rogers, Z.; Scb, S.; Evans, G.; Palanisami, T. Improved methodology to determine the fate and transport of microplastics in a secondary wastewater treatment plant. *Water Res.* **2020**, *173*, 115549. [CrossRef]
57. Conley, K.; Clum, A.; Deepe, J.; Lane, H.; Beckingham, B. Wastewater treatment plants as a source of microplastics to an urban estuary: Removal efficiencies and loading per capita over one year. *Water Res. X* **2019**, *3*, 100030. [CrossRef]
58. Lares, M.; Ncibi, M.C.; Sillanpää, M.; Sillanpää, M. Intercomparison study on commonly used methods to determine microplastics in wastewater and sludge samples. *Environ. Sci. Pollut. Res.* **2019**, *26*, 12109–12122. [CrossRef] [PubMed]
59. Liu, Y.; Wang, B.; Pileggi, V.; Chang, S. Methods to recover and characterize microplastics in wastewater treatment plants. *Case Stud. Chem. Environ. Eng.* **2022**, *5*, 100183. [CrossRef]

60. Hidalgo-Ruz, V.; Gutow, L.; Thompson, R.C.; Thiel, M. Microplastics in the Marine Environment: A Review of the Methods Used for Identification and Quantification. *Environ. Sci. Technol.* **2012**, *46*, 3060–3075. [CrossRef] [PubMed]
61. Song, Y.K.; Hong, S.H.; Jang, M.; Han, G.M.; Rani, M.; Lee, J.; Shim, W.J. A comparison of microscopic and spectroscopic identification methods for analysis of microplastics in environmental samples. *Mar. Pollut. Bull.* **2015**, *93*, 202–209. [CrossRef] [PubMed]
62. JRC, Joint Research Centre. Guidance on Monitoring of Marine Litter in European Seas. A Guidance Document within the Common Implementation Strategy for the Marine Strategy Framework Directive. MSFD Technical Subgroup on Marine Litter. 2013. Available online: https://op.europa.eu/en/publication-detail/-/publication/76da424f-8144-45c6-9c5b-78c6a5f69c5d/language-en (accessed on 9 July 2022).
63. Das, M. Man-Made Cellulose Fibre Reinforcements (MMCFR). In *Biocomposites for High-Performance Applications*; Woodhead Publishing: Sawston, UK, 2017; pp. 23–55.