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Feasibility of rapid gravity filtration and membrane ultrafiltration for the removal of microplastics and microlitter in sewage and wastewater from plastic industry

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ABSTRACT

Wastewater treatment plants (WWTPs) act as barriers in reducing uncontrolled microplastic and microlitter (MP-ML) emissions from both urban and industrial wastewaters. Despite removing most of the MP-ML, large quantities of this waste still enter the environment through WWTP effluents, which means further post-treatment technologies are needed. This study contains a technical evaluation of MP-ML removal from urban wastewater (UWW) and from the wastewater from the recycling plastic industry (PIWW) using two different pilot-scale post-treatment systems: rapid gravity filtration (RGF) and ultrafiltration (UF) membranes. The MP-ML mass concentrations contained in UWW and PIWW were measured by a simplified method adapted for the long-term monitoring of WWTP operations. The method was validated on standard samples. Despite the RGF system consumed less energy than UF treating UWW (0.097 kWh·m^{-3} and 0.156 kWh·m^{-3}, respectively), RGF was not efficient enough to properly decrease the risk of MP-ML emissions (39.5 ± 34.6 % of MP-ML removal). With respect to PIWW, the energy consumption of the UF plant decreased up to 0.059 kWh·m^{-3}. The combination of RGF and UF technologies was expected to reduce membrane fouling but it did not show significant differences in the mid-term operation.

1. Introduction

The worldwide concern about marine microplastics (MP) and microlitter (ML) has increased noticeably in recent years [1,2]. Microplastics are plastic particles smaller than 5 mm in any of their dimensions [3–5] and account for up to 90 % of total ML, which also includes other inorganic and organic materials such as metals, wood, rubber, glass and paper [6]. MP are considered by many scientists as pollutants of increasing concern due to their negative effects on the environment and wildlife, their durability in oceans, surface waters and sea sediments and their increasing production [7,8]. In this regard, small microplastics (and other microlitter) are easily ingested by fish, worms, shellfish, seabirds, turtles, etc., which remain in their gut for a long time, affecting their growth, development and reproduction [3,9,10]. Moreover, MP can adsorb and/or release certain toxic additives, resulting in increased concentration of toxic compounds that can cause carcinogenic or mutagenic effects in living organisms [11–14].

Between 1.15 and 2.41 Mt of plastics are believed to reach the oceans annually [15]. Around 80 % of them are produced by land-based activities and transported by winds, runoffs, infiltration, currents, uncontrolled disposals, etc., while the rest come from sea-based sources [1,3,13,16]. Many microplastic particles are found in urban and industrial wastewaters and thus have wastewater treatment plants (WWTPs) as the final barriers to reduce uncontrolled microplastic emissions into the environment [16–19]. Urban WWTPs normally remove microplastics and microlitter from wastewater streams with efficiencies over 90 % (Table 1), despite not being designed for that purpose [20,21]. However, some authors consider these removals insufficient [22]. Since

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WWTPs treat huge volumes of sewage, the particles remaining in their effluents (and emitted to the environment) can reach up to $1.5{\cdot}10^{10}$ MP·d^{-1} [2], which converts WWTPs not only into sinks but also into a source of microplastics [13,17,23,24] This also hinders the possibility of reusing wastewater for irrigation purposes due to the possible risks associated to their addition to croplands.

Additional technologies are thus needed to complement conventional wastewater treatment processes to boost microplastics and microlitter (MP-ML) removal [9,13,25]. Several authors have studied the physical removal of these pollutants by different post-treatment technologies such as rapid sand filtration, membrane systems and others (Table 1). By means of example, Talvitie et al. [21] compared rapid sand filtration and membrane bioreactor (MBR) technologies, obtaining 97 % and 99.9 % of MP removal, respectively. In other studies, MP removal efficiencies achieved in similar post-treatment systems vary widely depending on each case. For instance, for rapid sand filters, Bayo et al. [26] reported removal efficiencies of 75.5 %, while Wolff et al. [27] obtained much higher performance, i.e., 99.2 %–99.9 %. Similarly, studies based on membrane-based technologies such as microfiltration, ultrafiltration and MBR varied in a wide range of 79–99 % MP removal (Table 1).

It must be noted that the comparison between studies reported so far can be controversial considering the different characteristics of the wastewaters treated (Table 1), the different methodologies used for MP characterisation [16]. Furthermore, most of the studies developed so far about MP-ML removal in WWTPs are basically based on the characterisation of the microparticles in terms of number, size, shape, colour, etc. and on the analysis of influent and effluent concentrations of MP-ML [28–30]. Scarce information can be found about MP-ML mass concentration [31] and on the operability of these post-treatment systems and their mechanisms of removal [25]. It is well-known that in both rapid sand and membrane filtration, the primary removal mechanism is size exclusion, but adsorption due to electrostatic interactions or electrostatic repulsion between charged particles and membranes can also occur [32,33]. In the case of membranes, these interactions produce cake layers and pore blocking phenomena [33,34], which can result in higher fouling rates that can reduce filtration performance exponentially. The filtration process must be thus analysed carefully [25,36,37]. Apart from all the stated above, some questions about the role of post-treatment processes in MP-ML removal still remain unanswered [37]: Is the removal efficiency achieved in these systems enough to avoid the risks associated to MP-ML emissions to the environment? Which operating conditions are preferable to remove them? Are these post-treatment technologies energetically competitive with respect to the amount of MP-ML removed? This manuscript tries to find answers to these questions.

Apart from improving microplastic removal in urban WWTPs, reducing their emissions from other sources should be imperative to cope with this emerging problem [8,51]. In this respect, industrial wastewaters such as recycled plastic industry wastewater (PIWW) can also be a relevant source of microplastics that can influence the characteristics influent WWTP streams [39,52]. A comprehensive assessment of the removal (and/or possible recovery) of MP in PIWW worth being carried out in order to select the most appropriate remediation

Table 1
Results obtained in previous studies related to microlitter and microplastic removal in wastewater treatment plants (WWTPs).

Location	WWTP size (p.e)	Influent (MP·L^{-1})	Secondary effluent (MP·L^{-1})	Tertiary effluent (MP·L^{-1})	Post-treatment technologies	Removal efficiency (%)	Total discharge (MP·d^{-1})	Reference
Spain	210,000	12.43[a]	1.23	–	–	90.3	$6.7{\cdot}10^{6}$	[24]
Spain	29,777	4.40	–	0.92–1.08	MBR/RSF	75.5–79.0	$12.96{\cdot}10^{6}$	[26]
Israel	30,000[b]	64.78	2.72	1.97	Sand filtration + chlorination	97.0	$5.91{\cdot}10^{7}$	[38]
Spain	300,000	–	10.7	–	–	94	$3{\cdot}10^{8}$	[17]
Spain	82[b]	1567	131	–	–	92	$1.07{\cdot}10^{4}$	[41][e]
South Korea	20,840–26,545[b]	4200-5840	433–710	33–66	Ozonation/RSF	98.9–99.2	$8.8{\cdot}10^{8}$–$1.37{\cdot}10^{9}$	[40]
South Korea	469,249[b]	31,400	7863	297	Membrane disc filter	99.1	$1.39{\cdot}10^{11}$	[40]
South Korea	40,000	114–216	–	0.26–0.48	RSF + UV	99.8	$2.9{\cdot}10^{9}$	[37]
China	$3.1{\cdot}10^{6}$	126.0	37.9	30.6	UV	75.6	–	[41]
China	$3.5{\cdot}10^{6}$	1.57–13.69	0.20–1.73	–	–	79.3–97.8	≈$6.5{\cdot}10^{8}$	[42]
Finland	10,000[b]	57.6	1.0	0.4	MBR	98.3	–	[43][c]
Netherlands	9240–720,000[b]	68–910	51–81	–	–	72	–	[20]
USA	9900–$2.4{\cdot}10^{6}$	139	5.9	0.5–2.6	Sand/membrane filtration	97.2–99.4	$4.4{\cdot}10^{6}$–$1.48{\cdot}10^{10}$	[2]
Germany	7000-46,000	–	0.08–7.52	–	–	98	$4.19{\cdot}10^{4}$–$1.24{\cdot}10^{7}$	[4]
Scotland	650,000	15.7	0.25	–	–	98.4	–	[44]
Italy	80,000	3.6	0.76	0.52	UF	86	$4.15{\cdot}10^{7}$	[45]
Denmark	–	7216	54	–	–	99.3	3[d]	[18]
Thailand	130,000[b]	77	10.67	2.33	UF	96.97	$2.8{\cdot}10^{8}$	[46]
Finland	800,000	380–900	1.0–3.1	0.7–3.5	BAF	>99	$2.0{\cdot}10^{8}$–$7.9{\cdot}10^{8}$	[47]
Finland	Pilot-800,000	–	2.0	0.02–0.3	RSF/MBR/disc filter/ air flotation	95.0–99.9	$5.4{\cdot}10^{6}$–$8.1{\cdot}10^{7}$	[21]
Australia	$1.2{\cdot}10^{6}$	–	0.48	0.21–0.28	Sedimentation + UV/ UF/RO	92–99	$4.6{\cdot}10^{8}$	[23]
Turkey	87,500[b]	135.3	8.5	–	–	93.7	$5.25{\cdot}10^{8}$	[48]
Spain	70,417	3.78	–	1.38	RSF/UV	63.4	$1.6{\cdot}10^{7}$	[49]
Australia	13,000-320,000	840–3116[f]	–	7[f]	UF + UV disinfection	>99	–	[31]
Germany	32,000-725,000	–	0.7–19.6[f]	≈0–0.67[f]	RSF/MF/GAC/PAC	82.4–94	–	[50]
Germany	470,000-725,000	–	0.65–1.7	0.004–0.022	Sand filters	99.2–99.9	–	[27]

BAF: biologically active filter; GAC: granulated activated carbon; LOD: limit of detection; MBR: membrane bioreactor; MF: microfiltration; MP: microparticles; PAC: powdered activated carbon; RO: reverse osmosis; RSF: rapid sand filtration; UF: ultrafiltration; UV: ultraviolet disinfection.

[a] After grease removal.
[b] m^3·d^{-1}.
[c] Includes all textile fibres.
[d] t MP·y^{-1}.
[e] Industrial wastewater.
[f] MP concentration in μg·L^{-1}.

strategies. However, to the best of the authors' knowledge, the evaluation of post-treatment technologies for MP removal in PIWW regarding technical and feasibility factors has scarcely been done [27].

Another important issue related to the evaluation of microplastics removal lies on the fact that there are no standard methods for MP-ML measurement [4,30,31,53]. Furthermore, methods reported so far are commonly based on tedious MP visual counting and spectroscopic techniques that normally present high variability and can underestimate the number of MP-ML particles contained in the samples, especially in the smallest fractions [37,54,55]. Also considering that microplastics can reduce their size along the treatment units of WWTPs by multiple transformation processes such as mechanical and chemical abrasion, degradation, etc. [33], microplastic particle counting can produce some inconsistencies in the MP-ML evaluation. In addition, these methodologies are highly time-consuming, usually entailing manual separation of ML-MP particles, counting under an optical microscope and/or analysis by Fourier-transform infrared (FTIR) spectroscopy, taking several hours or even days [18,23,50]. From authors' experience, visual counting and FTIR analysis take an extra time (i.e., apart from the time needed for the pre-treatment of the sample described in Section 2.3.1) of around 12 h per sample. Depending on the number of particles present in the sample, it can be even higher. This complicates the monitoring and control of WWTP operations. To simplify the methodology and reduce uncertainties, MP-ML mass concentrations are expected to be more accurate, practical, and useful measurements for the long-term WWTP monitoring, as reported previously by other authors [31,50,52]. For these reasons, this study evaluates the MP-ML removal in rapid gravity filtration (RGF) and membrane ultrafiltration (UF) systems in terms of MP-ML mass concentrations.

The present study focuses on: i) quantifying MP-ML content in the effluent of a large-scale urban WWTP and recycled plastic industry effluents using a simplified methodology to quantify MP-ML mass concentrations; ii) analysing the MP-ML removal of two different pilot-scale post-treatment plants based on rapid sand filtration and hollow-fibre ultrafiltration membranes and their different behaviour treating UWW and PIWW; and iii) evaluating the operation and energy consumption of these post-treatment systems.

2. Material and methods

2.1. Characterisation of wastewaters

MP-ML removal was tested from: (a) urban wastewater (UWW); and (b) recycling plastic industry wastewater (PIWW). The UWW was obtained from the effluent of WWTP secondary treatment, although effluent from a Rapid Gravity Filtration (RGF) pilot plant (Section 2.2) was also evaluated. The PIWW was prepared simulating wastewater produced during polyethylene terephthalate (PIWW-PET) and high-density polyethylene (PIWW-HDPE) recycling. These plastics were selected since they are two of the most commonly used in the plastics industry (see their main characteristics in Table 2).

2.2. Wastewater treatment technologies

A rapid gravity filtration pilot plant and an ultrafiltration membrane pilot plant were used independently to treat the water effluent from a 50,800-p.e. WWTP in Valencia (Spain). The WWTP process is based on primary sedimentation and a conventional activated sludge (CAS) system with nitrification-denitrification. Apart from single-step post-treatments, the combination of the RGF plant and the UF plant was also evaluated. In a different treatment scheme, the UF plant was fed with PIWW-HDPE and PIWW-PET (Fig. 1).

2.2.1. Rapid gravity filtration (RGF) pilot plant

The RGF plant was composed of two 20-cm gravel layers with two different filtration media: silica sand (RGF-S) and spent activated carbon (RGF-C). The former is the typically used medium for polishing WWTP effluents, while the latter was tested to check whether this residue could be reused for removing microplastics from wastewater effluents. These media had a filtering area of 0.07 m^2 each, operating at a treatment flow rate in the range of 14.3–42.9 $m^3 \cdot m^{-2} \cdot h^{-1}$. The filtering flows were higher than those normally used in full-scale systems but allowed an evaluation of the limit filtration rate at which the system could operate. There were a number of nozzles at the bottom of the filter for back-flushing washing from an equalisation tank in a sequence of high-pressure air pulses (at 95 $Nm^3 \cdot m^{-2} \cdot h^{-1}$) and water (from the equalisation tank at 40–45 $Nm^3 \cdot m^{-2} \cdot h^{-1}$) and operated automatically when the head loss reached 80 % of the maximum.

2.2.2. Membrane ultrafiltration (UF) pilot plant

The UF plant mainly had two 2.5-m high membrane tanks (MT) with a working volume of 30 L. Both tanks contained a hollow-fibre bundle (KMS Puron® Koch Membrane Systems) with a membrane area of 3.4 m^2 and pore size of 0.03 μm. The plant also had a 50-L clean-in-place tank (CIP) to accumulate permeate, which was used to back-flush the membrane (BF). To reduce cake layer formation, air was sparged by a blower with a specific air demand (SAD) of 0.1–1.2 $Nm^3 \cdot m^{-2} \cdot h^{-1}$. The transmembrane flux operated by the membrane was in the range of 20–50 LMH, while the recirculation factor, i.e. the flow rate of the membrane feed with respect to the flow rate of the permeate was set at 2. The membrane combined the filtration–relaxation (F–R) stages and BF [56]. The F-R stages were fixed, lasting 150 and 30 s, respectively, while the membrane spent 60 s in BF every 5 F-R cycles. Eqs. S1–S7 were used to calculate the membrane parameters and the energy consumption of the UF plant.

2.3. Microplastic analysis

The methodology for the analyses was based on previously reported studies (see [16,18,23,24,43,57]) but simplified and modified to analyse MP-ML mass concentrations to evaluate the post-treatment technologies. In addition, some MP particle concentrations were done in UWW samples to characterise the urban wastewater.

2.3.1. Sample collection and pre-treatment

The sampling period was between June 2020 and March 2021. Grab samples were collected once every two weeks from the influent and effluent of the three different post-treatment schemes (Fig. 1). Following previous studies [16,47], large sample volumes of 150 ± 25 L were taken to obtain representative samples collected in-situ using the

Table 2
Average characteristics of municipal and industrial wastewaters treated.

	Wastewater	TSS ($mg \cdot L^{-1}$)	COD ($mg \cdot L^{-1}$)	Turbidity (NTU)	Conductivity ($\mu S \cdot cm^{-1}$)	pH
UWW	Secondary effluent	23 ± 7	78 ± 36	24.0 ± 3.0	1495 ± 221	8.2 ± 0.4
	RGF effluent	9 ± 3	33 ± 15	7.6 ± 0.8	1488 ± 125	8.1 ± 0.3
PIWW	HDPE	6 ± 2	n.d.	3.5 ± 0.6	1153 ± 145	8.3 ± 0.2
	PET	12 ± 2	n.d.	13.7 ± 1.9	1206 ± 136	8.2 ± 0.1

COD: chemical oxygen demand; HDPE: high-density polyethylene; n.d.: not detected; PET: polyethylene terephthalate; PIWW: plastic industry wastewater; TSS: Total suspended solids; UWW: urban wastewater.

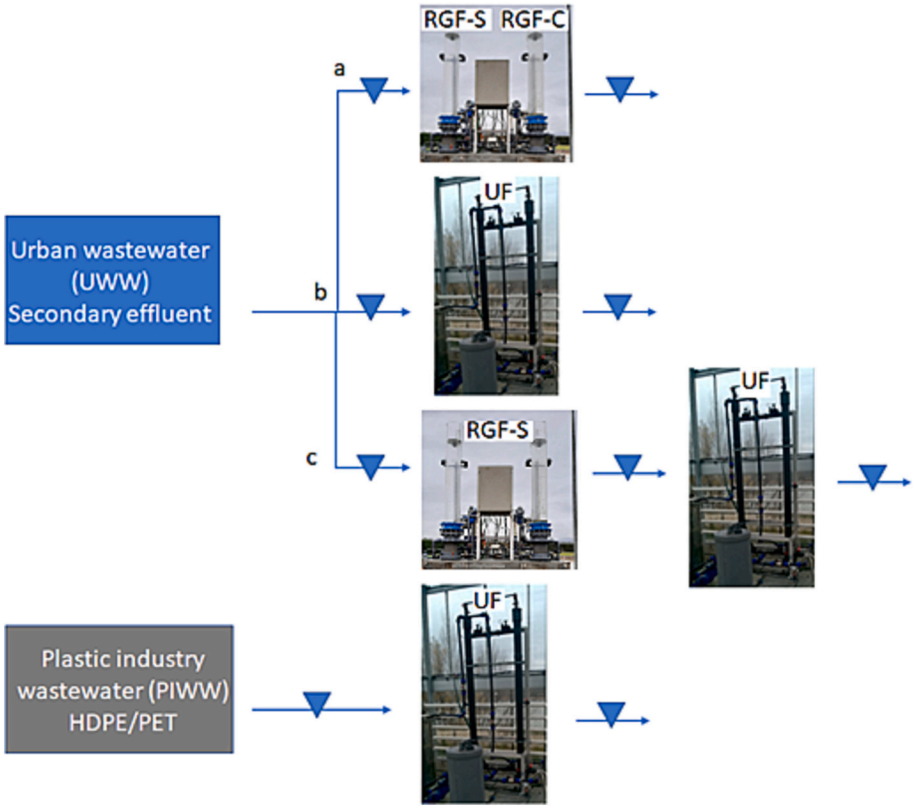

Fig. 1. Flow diagram of the pilot systems.
RGF: rapid gravity filtration; RGF-S: rapid gravity filtration with sand; RGF-C: rapid gravity filtration with activated carbon; UF: ultrafiltration plant.
▼: sampling point.

sampling device proposed in Ziajahromi et al. [23] (Fig. S1) that contained three stainless-steel grids (pore sizes: 500, 100, and 25 μm) which retained the MP-ML and fractioned them by size. The granulometries were selected according to their similarity to the microplastics found in wastewater [16,58]. It should be noted that mini-microplastics (MMP) and nanoplastics (NP) could not be measured in this way as they are smaller than 25 μm. The cover and bottom of the device were tightened by threaded studs to provide support and avoid wastewater leakage. A flowmeter was placed before the head of the sampling device to accurately measure the water volume sampled.

The grids (500, 100 and 25 μm) were then washed with 400–600 mL of distilled water to remove all the retained particles, after which the sample fractions contained in the glass flasks were oxidised by adding hydrogen peroxide (H_2O_2) (30 % *w/v*) and heated in a water bath to a temperature of around 65 °C, which enabled H_2O_2 evaporation, ensuring the degradation of all the organic matter while avoiding microplastic degradation [16]. This system was maintained for around 6–8 h, during which the water in the bath was maintained at the same level as the sample to avoid bubbles dispersing the microplastic particles. To confirm complete H_2O_2 removal, pH was monitored during heating, values close to 7 indicating that all the H_2O_2 had evaporated. NaCl (20 % *w/v*) was then added to increase the density of other inorganic fractions so that they could be separated from the MP by precipitation.

2.3.2. Samples determination

After pre-treatment, the liquid containing microplastics and other microlitter was filtered through stainless steel grids of different mesh sizes (which were narrower than the original filtering grids: 500, 100 and 25 μm) to retain the solid material and carry out a gravimetric analysis on each filter to determine MP-ML mass concentrations. To calculate this concentration, the difference between the final weight and the weight of the single filter was divided by the amount of wastewater filtered with the sampling device (Section 2.3.1). To avoid possible errors due to outer contamination, this concentration was corrected by subtracting the concentration of each corresponding blank sample.

Microplastic recovery tests were performed to validate the gravimetric method. A known amount of low-density polyethylene (LDPE) was added to deionised water to obtain standard samples with known MP concentrations. These pattern samples were processed by the pre-treatment described in Section 2.3.1. A gravimetric analysis was then made to determine the percentage of LDPE recovered. These tests were repeated using UWW as medium for the standard samples instead of deionised water, using blank samples to exclude the MP-ML in the medium recovered from the polymer addition. The results showed high MP recovery (frequently over 90 %) using both deionised water and UWW (Table S1), indicating that the method was appropriate for the study.

To check the percentage of MP according to the total ML, some microplastic particles that floated in the treated samples and were suctioned with a pipette, visually analysed and counted by means of a stereomicroscope (LEICA, LEITZ DM RX2). Their chemical composition was analysed by FTIR-Attenuated Total Reflectance (FTIR-ATR) (Fourier Perkin Elmer Frontier) to identify genuine MP particles.

3. Results and discussion

3.1. Characterisation of microplastics and microlitter

3.1.1. Urban wastewater (UWW)

MP-ML concentrations of each fraction of UWW samples can be seen in Table 3. The smallest size, i.e., in the range of 25–100 μm, was the most abundant fraction even though each particle is much lighter than those of bigger fractions. This probably occurs because the biggest fractions of MP-ML are removed more efficiently in the upstream WWTP

Table 3
Microplastic-microlitter content in UWW and PIWW.

Sample	Total MP-ML mass concentration (mg·L^{-1})		
	UWW	PIWW-HDPE	PIWW-PET
>500 μm	0.06 ± 0.07	0.93 ± 0.33	1.47 ± 0.22
100–500 μm	0.03 ± 0.02	0.82 ± 0.30	2.90 ± 0.42
25–100 μm	0.08 ± 0.06	4.04 ± 1.46	7.93 ± 1.16
Total (>25 μm)	**0.17 ± 0.08**	**5.8 ± 2.1**	**12.3 ± 1.8**

HDPE: high-density polyethylene MP-ML: microplastics and microlitter; PET: polyethylene terephthalate; PIWW: plastic industry wastewater; UWW: urban wastewater.

units and are also fragmented into smaller particles due to UWW processes [16,19,28]. From the samples analysed, high variability was obtained (high SD). This is widely observed in the literature since inaccuracies in the sampling and processing procedure of MP-ML detection are common and hard to be avoided [41,59,60].

From the microlitter particles tested by FTIR, 76 % were found to be microplastics. Fig. S2 shows the FTIR profiles obtained for one of the samples. Non-plastic microlitter (24 % of the particles analysed) was mainly composed of glass debris, natural fibres and recalcitrant organic matter. The MP-ML ratio was very similar to that obtained by Bayo et al. [49], who identified 72.41 % of the total microlitter as microplastics in wastewaters in Southern Spain. On the other hand, Gies et al. [61] found only 32.4 % of the total microlitter analysed from a WWTP in Vancouver (Canada) were actually microplastics. Of the MPs analysed by FTIR, 56.1 % were polypropylene (PP) and 43.9 % polyethylene (PE), some of the most common microplastic materials in wastewaters [3,9,13]. Although no other microplastic materials were tested, their presence in the samples could not be discarded, since the MPs that were not characterised by FTIR could have been from other plastic materials typically found in UWW, such as PET, PTFE (Teflon), polysterene (PS), PA, etc. [22,26,41].

3.1.2. Recycling plastic industry wastewater (PIWW)

PIWW from PET and HDPE recycling industries were also analysed. These wastewaters were less concentrated than UWW in terms of solids and turbidity and, unlike UWW, PIWW did not contain significant amounts of organic matter (Table 2). On the other hand, their MP mass concentration was higher than UWW, especially for the smallest fraction (Table 3). PIWW-PET and PIWW-HDPE showed similar particle size distribution, with most of the particles in the lowest size range, i.e. 64.5 % and 69.7 %, respectively. As aforementioned, the smallest fraction was also the most abundant (46.0 %) in UWW, but its distribution was significantly different (Fig. 2).

3.2. Microplastics removal

3.2.1. RGF plant

In the RGF-S plant, MP-ML contained in UWW were caught in grains of sand or adhered to the particles' surface, reducing MP concentration in the treated water by 39.5 ± 34.6 % in comparison to RGF-S influent. The rest of MP-ML, which were probably smaller than the gaps between grains, had the potential to pass through the system [22,40]. As the MP-ML removal achieved in pre-treatment, primary and secondary treatments is normally high, i.e. around 95–97 % [16,19,22], the extra MP-ML removal achieved in the RGF-S system could be considered insufficient and would highlight the need to look for other technologies that successfully remove all types of MPs. However, further research is needed to properly assess the risks that RGF-S effluents could present when emitted to the environment or to croplands (if wastewater is reused for irrigation) since the consequences of emitting these pollutants after tertiary treatment are unclear [52].

With respect to the RGF-C system, although microplastics cannot be adsorbed to activated carbon, partial MP-ML removal by size exclusion could be expected due to the small size of activated carbon granules. However, no significant MP-ML removal was observed, which confirmed that spent activated carbon is not a good compound to remove microplastics, so it cannot be reused for this application. For this reason, RGF-C was not further evaluated in the following sections. From the particles analysed, it was observed that most of the MP-ML in RGF-S effluents were fibres, i.e., 70 %. This is in line with previous studies that found the dominance of microfibres in effluents from secondary and tertiary treatments due to their high length-width ratio [62,63].

3.2.2. UF plant

In none of the UWW and PIWW samples analysed in UF effluents were measured higher MP-ML concentrations than in blank samples (for all particles sizes). It was thus assumed that the UF plant was able to efficiently remove all the particles larger than 25 μm, suggesting that UF is a good option to remove microplastics higher than this pore size from different types of wastewater. However, particles with sizes smaller than the membrane's pore (0.03 μm) such as MMP and NP could still be present in the permeate without having been measured [33,64,65]. Other authors have reported the presence of microplastics larger than 25 μm (mainly fibres) in membrane effluents [23,37,43,45,66].

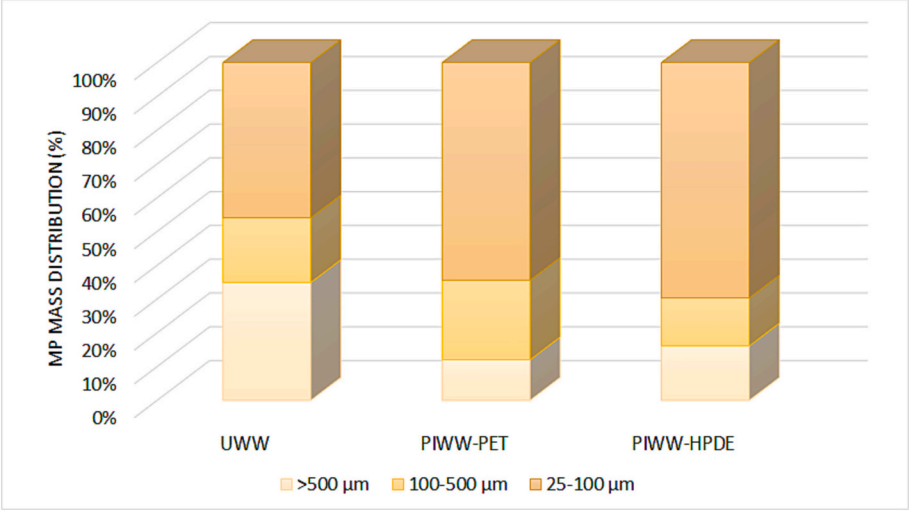

Fig. 2. Particle size distribution of wastewaters; a) UWW:urban wastewater; b) PIWW-PET: recycling plastic industry wastewater-PET; c) PIWW-HDPE: recycling plastic industry wastewater-HDPE.

However, it is difficult to verify whether these particles were actually present in the effluent of their membrane systems, appeared during the manipulation of the sample or were due to malfunctions of the membrane filtration such as small leaks between seals, breakage of membranes or to partial contamination due to the pipelines or storage tanks [29,46,61]. Since no MP-ML were detected, the membranes from the UF plant were considered to operate correctly, without malfunctions. However, further research is needed to clarify the mechanisms of membrane filtration in this process. To sum up, the results obtained in this study confirmed the higher efficiency of UF membranes than RGF for removing MP-ML from UWW, in disagreement with Bayo et al. [26], who found no significant differences between both systems.

3.3. Evaluation of pilot-scale post-treatments

3.3.1. RGF-S plant

For all the conditions tested, the RGF-S plant showed no significant differences in filtering time. It should be noted that the reduced permeability of the granular media (measured as a pressure drop) was due to the accumulation of suspended solids from the UWW. No correlation was found between pressure drop and MP concentration, probably due to their low amounts [37,42]. On the other hand, the washing conditions tested were considered effective to recover the system's filtering capacity, since the head loss was always <15 % of the initial after the backflush. This step was applied every 3–5 days, while other authors have reported regular back-flush cleaning every 2–3 days for RFG systems [67]. It can be thus concluded that this system is technically feasible to remove certain MP-ML fractions, although their removal was not as high as expected (Section 3.2.1).

With respect to energy consumption, 0.031 and 0.020 kWh m^{-3} were obtained for pumping during feeding and backflushing steps, respectively, while it reached 0.046 kWh m^{-3} for blowing. Total consumption of the RGF-S plant was thus a maximum of 0.097 kWh m^{-3}.

3.3.2. UF plant

To maintain membrane fouling in low rates, membrane systems are often operated under sub-critical conditions [68]; i.e., at transmembrane fluxes (J) that are under the critical permeate flux (J_c). J_c is defined as the flux at which the transmembrane pressure (TMP) and J are not linearly related [69], so that it can serve as an indicator of the system's filtering capacity.

Short-term tests were carried out in the UF plant to obtain J_c. [69]. As can be seen in Fig. 3, J_c was not reached in either UWW or PIWW since TMP increased linearly with respect to J with no shift in their trends. This was confirmed by the high R^2 values of these lines, which were over 0.97 in all cases (Table S2), suggesting that these permeate flux ranges (under 50 LMH) were appropriate for continuous operations.

Despite the high membrane fluxes tested, the filtration resistances obtained (5.42–12.16·10^{11} m^{-1}) were relatively low in comparison to other studies. For instance, Robles et al. [70] reported R_{20} values one order of magnitude higher during the continuous ultrafiltration of an anaerobic culture operating at transmembrane fluxes of 10–13.3 LMH. However, in that study the solids concentration was much higher (in terms of g·L^{-1}), while the wastewaters tested in the present study only contained 6–25 mgTSS·L^{-1} (Table 2). This means that the wastewaters tested in this study could be filtered at much higher transmembrane fluxes, which would reduce the CAPEX associated with the membrane surface and the OPEX associated to membrane replacement [71].

Of the wastewaters tested in this study, UWW showed significantly higher R_{20} values (p-value < 0.05) than the others (Table S2), despite having lower microplastic concentration than PIWW (Table 3). This was probably due to the higher solid and organic matter concentrations of UWW than PIWW [72] (see Table 2). For UWW, membrane resistance was commonly higher at decreasing SAD since the goal of air sparging during filtration is to reduce cake layer formation due to the accumulation of solids (reversible fouling). In consequence, the higher the SAD, the smaller the cake layer and the lower the resistance to filtration [73]. However, SAD has to be optimised since it is usually the main energy consumer in ultrafiltration systems [74]. In this study 0.4 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ was considered the most appropriate SAD from those tested, thus it was used to calculate the energy consumption of UF membranes, together with a J_{20} of 40 LMH since at this permeate flux the TMP was lower than 150 mbar for all the wastewaters tested (Fig. 3). Under these conditions, the consumption of the UF system during continuous operation

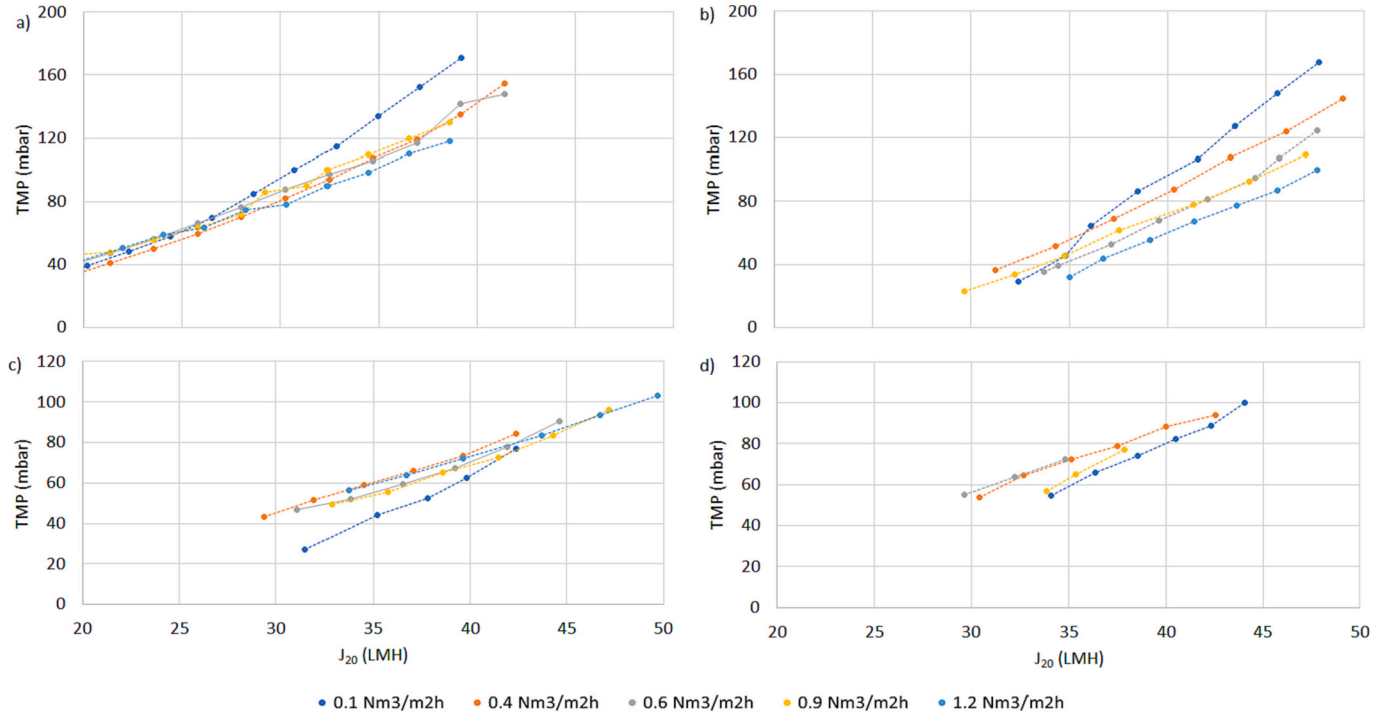

Fig. 3. Results of critical flux tests obtained under different specific air demand (SAD) for: a) UWW; b) RGF effluent; c) PIWW-PET; d) PIWW-HDPE.

accounted for 0.156 kWh·m^{-3}, 83 % of which was for air scouring. Considering that a classical activated sludge (CAS) system (with nitrification-denitrification) consumes around 0.3–1.4 kWh·m^{-3} [75], adding a UF system would require a total consumption of 0.46–1.56 kWh·m^{-3}, which is in the lower range of the power needs of MBR systems, i.e. 0.5–2.5 kWh·m^{-3} [75]. Moreover, membrane life is expected to be much higher in a UF than MBR system filtering secondary effluents, due to the large amounts of suspended solids present in MBRs, i.e. up to 13,500 mgTSS·L^{-1} [76]. It should also be noted that UF processes would remove not only MPs from UWW, but also other macro and micro-pollutants such as suspended solids and pathogens [77]. This makes UF a potential technology for water reuse, but in this case, these pollutants should be monitored to comply with EU Regulation 741/2020 [78].

In PIWW, resistance to filtration did not significantly vary with SAD (Table S2). This suggests that membrane resistance in PIWW was not significantly influenced by cake layer formation but was influenced by partial pore blocking, probably due to the absence of organic matter in PIWW (Table 2). In fact, organic particles in UWW have been reported to increase membrane fouling, especially when the cake layer on the membrane surface is being built up, i.e., when membrane resistance is low [79], as in this case. Increasing SAD can thus be an efficient way of reducing membrane fouling only when the wastewater filtered contains organic matter but seems to be inefficient for wastewaters mainly containing inert particles. For this reason, the lowest SAD for PIWW was selected, i.e., 0.1 Nm^3·m^{-2}·h^{-1}, with a total energy consumption of 0.059 kWh·m^{-3}, 54 % of which was for air sparging. This consumption is 62 % lower than that of UWW, suggesting that the UF plant is a more cost-effective system of filtering PIWW than UWW. In addition, the MP-concentrated stream in PIWW could be assessed for the recovery of plastic materials for recycling, thus improving the circularity of the process.

3.3.3. *Combination of RGF-S and UF*

The results of the present study indicate that combining both technologies operated under the same conditions would increase the total energy consumption to 0.254 kWh·m^{-3}. The goal of this treatment scheme was to check whether the RGF-S system would reduce UF membrane fouling significantly to compensate for the additional costs of having two post-treatment steps. Membrane life could also be expected to extend when filtering RGF-S effluent instead of UWW, due to its lower resistance to filtration (Table S2).

To verify this point, continuous filtration of both UWW and RGF-S effluent was carried out in the same operating conditions for one month (Fig. 4). TMP evolution, an indicator of the membrane fouling rate, was similar in both wastewaters, even though the RGF-S showed significantly lower solids and organic matter concentrations (Table 2) and short-term resistance to filtration (Table S2) than UWW. It should be noted that despite periodic backflushing (Section 2.2.2), MP-ML can stick to the membrane and can be hard to remove [36]. Also, the RGF-S system reduced not only the concentration of solids from 23 to 15 mg TSS·L^{-1} but also the average size of the solid particles; while in UWW the largest number of particles were in the 79–91 μm range, in the RGF-S effluent the most abundant particles were between 52 and 60 μm (Fig. S3). It is therefore possible that the smaller particle size in the RGF-S effluent caused more severe pore blocking and a less porous cake layer that hindered permeability [80], with a comparable medium-term (Fig. 4) and different short-term fouling rate to UWW (Fig. 3). Hence, although both UF and the combination of RGF-S and UF obtained the same MP-ML removal efficiency, the combination did not appear to be an attractive option for UWW post-treatment, as it would entail higher CAPEX and OPEX than UF only and did not reduce the mid-term average fouling rate. Post-treatment by UF thus appears to be the best option, although it should be optimised to reduce its high energy consumption for UWW of 0.156 kWh·m^{-3}.

Finally, it must be highlighted that the control of MP-ML emissions must be targeted by the avoidance of massive and indiscriminate use of plastic in order to reduce MP-ML discharges from WWTPs, especially if wastewater wants to be reused to implement circularity in the water sector [5,81]. In this respect, it must be noted that MP removal would be associated to the removal of other contaminants of emerging concern that can be part of microplastic composition or being attached to their surface [13,60].

4. Conclusions

This study assessed RGF and UF technologies for the removal of MP-ML from UWW and PIWW. The methodology used in this study was

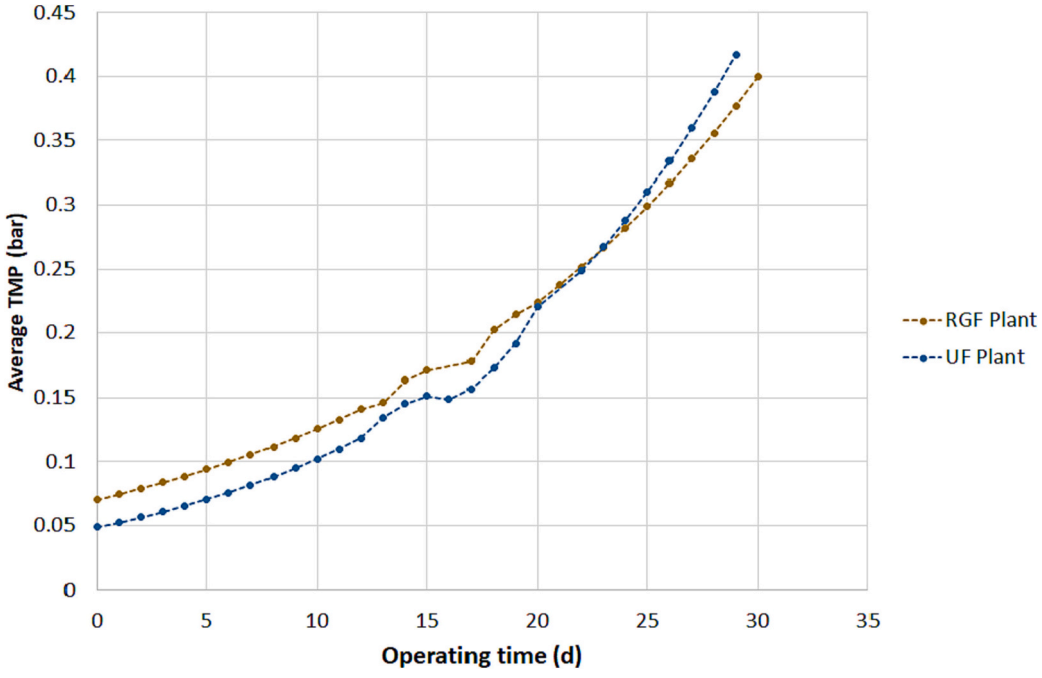

Fig. 4. Evolution of the daily average transmembrane pressure (TMP) of the UF plant during mid-term operation.

based on mass concentration instead of the commonly used particle concentration since the latter implies higher uncertainties and is more time consuming, hindering the application of these methodologies to WWTP monitoring. This method was validated on standard samples. The RGF-S system removed 39.5 ± 34.6 % of MP-ML in UWW (RGF-S influent), whereas no MP-ML were detected in UF effluents. Hence, even though the RGF-S system was technically feasible and consumed less energy than UF, i.e., 0.097 kWh·m^{-3} and 0.156 kWh·m^{-3}, respectively, RGF-S was not efficient enough to properly decrease the risk of MP-ML emissions. With respect to PIWW, the energy consumption of the UF plant decreased up to 0.059 kWh·m^{-3} due to the lower membrane fouling rates needed to operate the system, which was probably related to the negligible amounts of organic matter in PIWW. UF technology thus seems to be appropriate for microplastic removal from this type of wastewater. The combination of RGF-S and UF technologies was expected to reduce membrane fouling but it did not show significant differences in the mid-term operation. The UF membrane system thus appeared as the most appropriate system to reduce MP-ML, but further research is needed to optimise its operation since the energy consumption due to air sparging remained high when treating UWW.

Finally, the control of MP-ML emissions by the avoidance of massive and indiscriminate use of plastic should be the main strategy to reduce MP-ML discharges from WWTPs, especially if wastewater wants to be reused.

Declaration of competing interest

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

Data availability

The authors do not have permission to share data.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jwpe.2022.103452.

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