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### ► To cite this version:

J. Yang, M. Monnot, Y. Sun, L. Asia, P. Wong-Wah-Chung, et al.. Microplastics in different water samples (seawater, freshwater, and wastewater): Removal efficiency of membrane treatment processes. *Water Research*, 2023, 232, pp.119673. 10.1016/j.watres.2023.119673. hal-03989908

**HAL Id: hal-03989908**

**<https://hal.science/hal-03989908v1>**

Submitted on 16 Feb 2024

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524 **Microplastics in different water samples (seawater, fresh water and**  
525 **wastewater) - Part II: Removal efficiency of membrane treatment processes**

526  
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532 **Abstract:**

533 Microplastics (0.1  $\mu\text{m}$ –5 mm) as a newly emerging pollutant have attracted increasing global  
534 attention due to their widespread distribution and various adverse effects on natural environments  
535 and human health potentially. However, research in the field is still in the early stages of  
536 exploration as there are almost no relevant standards or policies made for the detection, emission  
537 and removal methods for microplastics in any country or region. Therefore, this study used an  
538 improved quantitative and qualitative analytical methodology for microplastics detection by  $\mu$ -  
539 FTIR spectroscopy carried out with siMPle analytical software and explained in Part I. In this part  
540 II, the removal efficiency of membrane treatment processes is evaluated using this methodology.  
541 The presence of microplastics was measured in different water sources and their treatment plants  
542 (seawater, three municipal and a pharmaceutical wastewaters and three drinking waters) with the  
543 same analytical method. Polyethylene is the dominant material in almost all water types followed  
544 by polypropylene, polystyrene, and polyethylene terephthalate. Almost all treatment technologies  
545 in water treatment plants can remove microplastics whatever the feed water type and concentration  
546 of microplastics. But some treatment processes or transport pipes could also cause additional  
547 pollution of the water with microplastics. Moreover, ultrafiltration was confirmed to be an  
548 excellent process for microplastic removal both in industrial and lab-scale experiments. The  
549 200 kDa ultrafiltration membrane could retain 70–100% and 80–100% of microplastics in quantity  
550 and in surface area, respectively. Thus, ultrafiltration leads to a considerable removal efficiency  
551 on microplastics and it also can greatly control the dimensions of microplastics in smaller ranges.

552

553 **List of abbreviations**

554

<b>Abbreviation</b>	<b>Full name</b>
ABS	Acrylonitrile Butadiene Styrene
ATR	Attenuated total reflection
DWTP	Drinking water treatment plant
EPS	Expandable Polystyrene
EVA	Ethylene Vinyl Acetate
FTIR	Fourier-transform infrared
MCT	Mercuric cadmium telluride
MP	Microplastic particle
PA	Polyamide (nylon)
PAN	Polyacrylonitrile
PC	Polycarbonate
PE	Polyethylene
PES	Polyether Sulfone
PET	Polyethylene Terephthalate
POM	Polyoxymethylene (Acetal)
PP	Polypropylene
PS	Polystyrene
PTFE	Polytetrafluoroethylene
PUR	Polyurethane
PVA	Polyvinyl Alcohol (sometimes Polyvinyl Acetate)
PVC	Polyvinyl Chloride
SI	Supplementary Information
SWTP	Seawater treatment plant
TOC	Total organic carbon
TP	Total particles
UF	Ultrafiltration
UP	Ultrapure
WWTP	Wastewater treatment plant

555

556

## 558 1. Introduction

559 Rapid urbanization and industrialization promoted the development and progress of society,  
560 while bringing more potentially emerging contamination risks, such as microplastics (0.1 $\mu$ m ~5  
561 mm) (Drummond et al., 2022). Public concerns on microplastic transport and transfer, ecological  
562 risks, and chronic toxicity to living, were increased globally. Excessive production of plastics has  
563 risen over 20 fold over the last half century and is predicted to worsen at least 2050 (Kahane-  
564 Rapport et al., 2022). The widespread applications and improper disposal contributed to the  
565 ubiquitous existence of plastic debris in diverse aquatic systems, such as wastewater, marine water,  
566 freshwater, etc. (Luo et al., 2019). Specifically, 95-99% of MPs derived from discarded plastics  
567 could be enrolled into raw wastewater and wastewater treatment plants (WWTPs) were the major  
568 receptors for microplastics derived from industries, landfill, domestic wastewater, and stormwater  
569 (Wu et al., 2022). The distribution and abundance of microplastic particles (MPs) in WWTPs were  
570 greatly related to human activities and urban agglomeration (Chen et al., 2020). Abundance of  
571 MPs in marine waters was demonstrated to range from tens to thousands of particles per cubic  
572 meter, and most marine organisms were contaminated with several to hundreds of MPs per  
573 individual (Bakir et al., 2020). Even in freshwater, MPs of polypropylene (PP), polyethylene (PE),  
574 polystyrene (PS), and polyethylene terephthalate (PET) were frequently detectable, accounting for  
575 70% of total MPs (Li et al., 2020). Additionally, the rivers, lakes, and even glaciers were all  
576 contaminated with microplastics. For instance, MPs in Lake Geneva (Europe) surprisingly reached  
577 a concentration of 48,146 MP $\cdot$ km<sup>-2</sup> (Faure et al., 2012) and the annual deposition flux of MPs in  
578 Demula glacier (Asia) was evaluated to 7640  $\pm$  720 to 9550  $\pm$  900 MP $\cdot$ m<sup>-2</sup> (Wang et al., 2022).

579 In view of environmental risks, the presence of microplastics increased its toxicity via  
580 compound contaminations and posed multi threats to humans and other living beings.  
581 Microplastics most with hydrophobic surfaces probably act as carriers for toxic  
582 organics/inorganics adsorption and for microorganism growth (Yang et al., 2022). More studies  
583 illustrated that humans may be exposed to MPs through inhalation, ingestion, and dermal contact,  
584 and MPs could be transported via blood (Huang et al., 2022; Na et al., 2021). Based on extensive  
585 investigations, large quantity and widespread of microplastic contaminations need to be furtherly

586 researched and demand prompt solutions. While, no specific treatment technology on MPs  
587 removal has been determined currently, it is necessary to explore the fate of MPs in water treatment  
588 plants and investigate the effective treatment technology on MPs removal. Additionally, the  
589 distribution of MPs in concentration, MP types, and MPs dimensions varied a lot even in same  
590 water types (Bretas Alvim et al., 2020; Simon et al., 2018), as shown in Table S1 of supplementary  
591 information (SI) with data of MPs in different water types from literatures. Different studies  
592 showed poor comparability because of the different detection methods and operation processes.  
593 Previous studies were mainly focused on individual stages in the assessment of MPs, while an  
594 analysis of all stages (i.e., sample extraction, processing, identification, quantification, and  
595 transportation) was required for a comprehensive understanding of the contamination caused by  
596 MPs. Therefore, it should be firstly considered to choose a suitable detection method of MPs with  
597 high efficiency and high accuracy to investigate the distribution and transportation of MPs.

598 In France, the occurrence of MPs in natural aqueous environments was rarely mentioned, and  
599 the removal of MPs through treatment plants remains to be explored. Therefore, this study was  
600 designed to investigate the distribution of MPs in France with two main objectives: (1) To obtain  
601 and compare the essential information on distribution and abundance of MPs from wastewater,  
602 seawater, surface water, and underground water; (2) To find out the footprint of MPs in different  
603 water treatment plants and calculate the related removal efficiency by each treatment process. To  
604 be more precise, the reliable analytical method on MPs identification and quantification by  $\mu$ -FTIR  
605 with siMPle software proposed in Part I was adopted in this Part II. A three-item characterization  
606 of MPs (counts, dimensions or surface area, types) is possible with this method. Samples from  
607 wastewater treatment plants (WWTPs), seawater treatment plants (SWTPs), and drinking water  
608 treatment plants (DWTPs) were all investigated with the same analytical method to evaluate MPs  
609 removal by primary, secondary, or tertiary treatment processes. Moreover, the treatment by  
610 ultrafiltration (UF) membranes was particularly assessed. This study gives information on  
611 microplastics contamination in France with a unified analytical method and in different water  
612 samples.

## 613 2. Material and Methods

### 614 2.1. Sample Sources and pretreatment

615 Three types of water (Table 1) were analyzed in this study: (i) samples from a SWTP equipped  
 616 to treat seawater after shellfish farms and before discharge into the ocean with two main treatment  
 617 routes, as shown in Figure 1. Particularly, in treatment route 2, two groups of 200 kDa UF modules  
 618 were located in different positions: semi-industrial scale UF with more than 4 years of operation  
 619 (membrane area 9 m<sup>2</sup>- Old UF) and industrial scale UF with less than 1 month operation  
 620 (membrane area 240 m<sup>2</sup>- New UF).; (ii) samples from four different WWTPs sites; and (iii)  
 621 samples from three drinking water treatment plants (DWTPs). To avoid external pollution, all  
 622 samples were collected and transported in 1 L pre-cleaned glass bottles with 3 replicates and frozen  
 623 up at -20C° before analysis. The bottles were closed immediately after sampling. The contact with  
 624 the bottleneck and inner covers was prevented during sampling. .

625 The pretreatment of membrane process was mainly described in Part I. The chemicals of 30%  
 626 (w/v) H<sub>2</sub>O<sub>2</sub>, 10% (w/v) KOH, 1–10% H<sub>2</sub>SO<sub>4</sub>, and Fenton reagent were applied for digestion under  
 627 room temperature, mainly based on the components of samples. Ultrapure water and filtrated  
 628 ethanol were used for rinsing and cleaning. The plastic-made devices or containers were avoided,  
 629 and the glass devices or containers were pre-cleaned with selected rinsing solution (whose positive  
 630 control was made) before using. MPs extraction was operated in cleaned fume hood, with white  
 631 lab coats worn at all times.

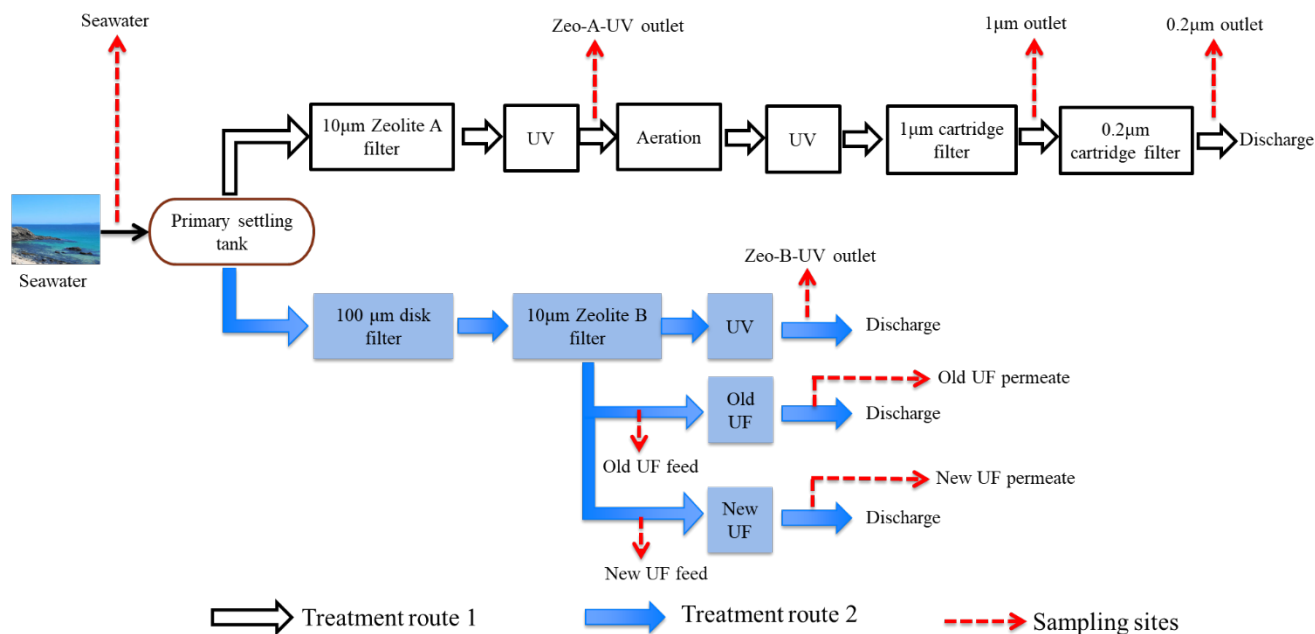
632 Table 1 Information of water treatment plants

Types	Names	Samples	Main process	Location in France (French department)	Sampling date
Wastewater	WWTP 1	Pharmaceutical membrane bioreactor (MBR) feed and effluent	MBR (0.02 µm UF)	Alpes-de-Haute-Provence	April 2021
	WWTP 2	Municipal wastewater and secondary effluents	MBR (0.1 µm UF)	Bouches-du-Rhône	October 2021
	WWTP 3		Physico-chemical with biofiltration	Bouches-du-Rhône	October 2021
	WWTP 4		Activated sludge, 200 kDa UF	Bouches-du-Rhône	October 2021
Seawater	SWTP	Seawater and secondary/tertiary effluents	Zeolite, filters, 200 kDa UF	Vendée	April 2021
	DWTP 1	Underground water and effluents	Sieves, 200 kDa UF	Paris	September 2021

Drinking water	DWTP 2	Surface water and effluents	200 kDa UF	Bouches-du-Rhône	October 2021
	DWTP 3	Surface water and effluents	Sedimentation, sand filtration	Bouches-du-Rhône	October 2021

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636

637

Figure 1 Seawater treatment plant and sampling locations

## 638 2.2. Analytical Method (full details in Part I)

639 The detection of MPs was achieved by the  $\mu$ -FTIR microscope (Perkin Elmer, USA) coupled  
 640 with siMPle software (Germany), as previously described in Part I. The turbidity of samples was  
 641 tested in the laboratory with a turbidity meter, Turb 550 IR (WTW ,Germany). A TOC-L  
 642 instrument (Shimadzu, Japan) based on the 680 °C catalytic oxidation method was used to measure  
 643 the concentration of total organic carbon (TOC).

644 The retention efficiency (by membranes) or removal efficiency (by other treatments) was  
 645 determined as the decreased MPs concentration compared to MPs concentration in influent of  
 646 related treatment process, shown as Eq. (1):

$$647 E_{\text{retention/removal}} = (1 - C_{\text{MPs\_feed}} / C_{\text{MPs\_effluent}}) \times 100\% \quad \text{Eq. (1)}$$

648 Where  $C_{\text{MPs\_feed}}$  represented MPs concentration before treatment;  $C_{\text{MPs\_effluent}}$  represented the  
649 MPs concentration after treatment. This equation was applied for MPs concentration in quantity  
650 and surface area.

### 651 2.3. Sample Pretreatment

652 The digestion methods used in this study included 30% (w/v)  $\text{H}_2\text{O}_2$  (Fisher Chemical), 10%  
653 (w/v) KOH (Fisher Chemical), 1–10%  $\text{H}_2\text{SO}_4$  (Fisher Scientific), and Fenton reaction. The  
654 selection of digestion was mainly based on compositions of samples in terms of organic matters  
655 and suspended solids. Samples with digestion requirements was firstly digested with  $\text{H}_2\text{O}_2$  at  
656 volume ratio ( $V_{\text{sample}}:V_{\text{H}_2\text{O}_2}$ ) from 1:1 to 1:2 for 1–14 days. When 14d of  $\text{H}_2\text{O}_2$  was not available,  
657 Fenton reagent (0.05M (II)  $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$  with 30%  $\text{H}_2\text{O}_2$  at volume ratio of 1:1) for stronger  
658 oxidation were applied. Particularly, seawater samples needed digestion with 10% (w/v) KOH  
659 within 24 h, and 1-10%  $\text{H}_2\text{SO}_4$  was used to adjust pH and eliminate precipitation. The samples  
660 were digested to clearer visible survey by avoiding occlusion by organic matters. During digestion,  
661 the glass bottle was covered with aluminum foil to prevent airborne contamination. All the  
662 digestion processes were operated under room temperature. Ultrapure water and filtrated ethanol  
663 was used for rinsing throughout the whole experiments. Three types of filters, 25  $\mu\text{m}$  stain steel  
664 filter and 3 or 5  $\mu\text{m}$  gold filter (i3 TrackPor P, Germany), were used for MPs extraction before  $\mu$ -  
665 FTIR imaging by a vacuum Büchner funnel apparatus (Part I). The MPs abundance in chemicals  
666 were assessed before experiments with ignorable influences on final results.

667 More detailed information on sampling, digestion, rinsing process,  $\mu$ -FTIR and siMPle, and  
668 operational conditions can be found in Part I.

### 669 2.4. Lab-scale Filtration of PE MPs by UF membranes

670 UF membranes were multichannel hollow fibers made of polyether sulfone (PES)  
671 (ALTEON™ I, SUEZ Aquasource®, France). The molecular weight cut-off (MWCO) of UF  
672 membrane was 200 kDa, with pore size of 20 nm. A UF module was built with one fiber which  
673 was composed of 7 channels with inner diameter of 0.9 mm, and installed into a PTFE external  
674 shell plugged with epoxy glue on both sides of the module, allowing in/out dead-end filtration. The  
675 module was 20 cm in length resulting in approximately a specific surface of  $3.96 \times 10^{-3} \text{ m}^2$  and an

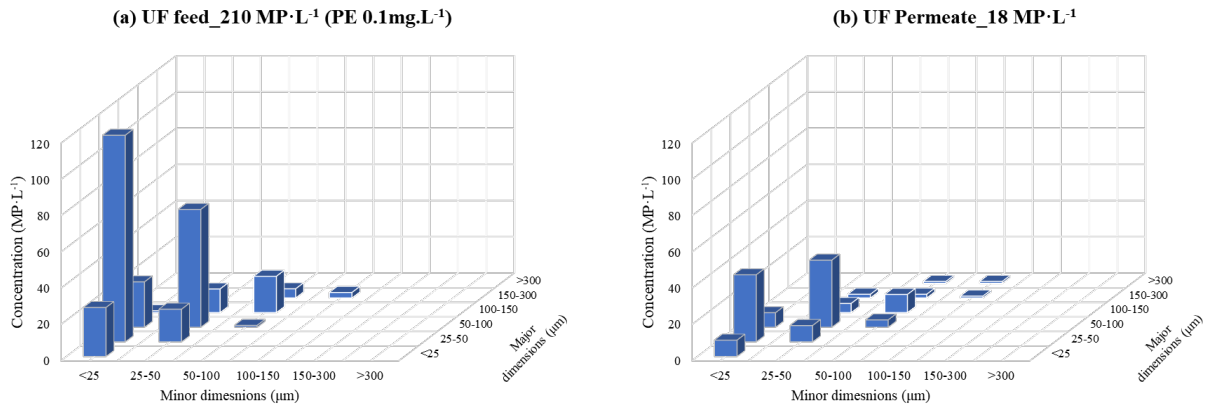


676 concentration volume of 0.89 mL. Polyethylene (PE) microspheres (10-150  $\mu\text{m}$ , Cospheric, USA)  
677 were used to prepare 0.1  $\text{mg}\cdot\text{L}^{-1}$  PE suspension as UF feed. The dead-end filtration test (once for  
678 1 L) was operated at a constant transmembrane pressure (TMP) of 0.3 bar. The UF system (pipes,  
679 connections, or containers) contained none PE particle. Before filtration, UF membranes were  
680 rinsed with 1 L of ultrapure water ( $300\text{ L}\cdot\text{m}^{-2}$ ) under a TMP of  $1.0 \pm 0.1$  bar to remove the  
681 preservative agent (glycerol). UF permeate was collected in pre-cleaned glass bottles and  
682 immediately analyzed by the developed method in Part I.

### 683 3. Results

#### 684 3.1. Microplastics Retention by UF membrane in Lab-scale

685 The investigation of UF membrane on PE MPs retention in lab-scale was operated, as shown  
686 in Figure 2. By identification, PE was the main type of MPs in  $0.1\text{ mg}\cdot\text{L}^{-1}$  PE synthetic suspensions,  
687 shown in SI, Figure S1. To be noted, the only MPs considered in this part were PE MPs for  
688 calculations to avoid the influences of airborne contamination and other secondary contamination  
689 on final results. The 200 kDa UF membrane achieved 91% retention of MPs in quantity, resulting  
690 in  $18\text{ MP}\cdot\text{L}^{-1}$  in UF permeate. The retention rate of microplastics in surface area could reach more  
691 than 98%. After all, the lab scale study confirmed that UF membrane could effectively retain MPs  
692 from inlet water and control the dimension of PE MPs in smaller ranges (minor dimensions ( $d_{\text{min}}$ )  
693  $<50\text{ }\mu\text{m}$  and major dimensions ( $d_{\text{maj}}$ )  $<150\text{ }\mu\text{m}$ ), but still some PE MPs with a larger dimension  
694 than UF pore size ( $0.02\text{ }\mu\text{m}$ ) were found in permeate. The question was opened to know where  
695 these particles come from knowing that membrane integrity was checked: airborne contamination,  
696 UF plastic module and permeate pipe degradation, more porous module cross section, etc. UF  
697 efficiency for MP removal at full scale will also be assessed in the following sections.

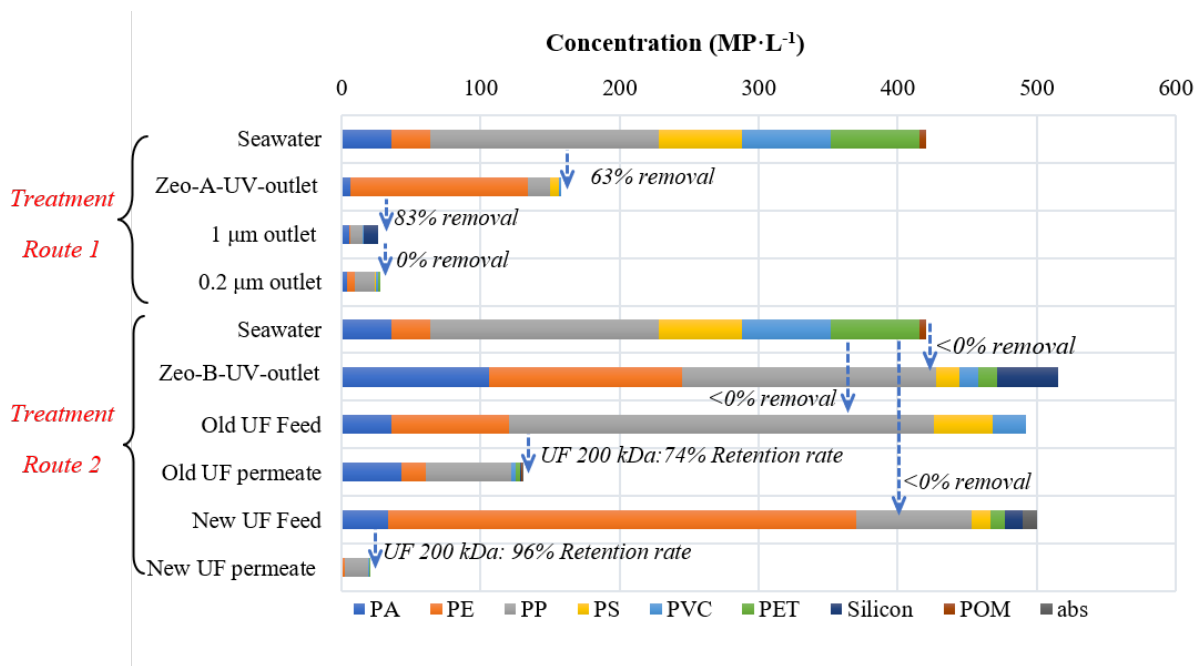


698

699 Figure 2 Dimensions and Concentration Distribution of PE MPs in (a) UF feed (before filtration)  
 700 and (b) UF permeate (after filtration).

701 3.2. Microplastics Retention in Seawater Treatment Plant

702 The publications on the presence of MPs and its related hazardous in marine environment  
 703 emerged exponentially, but there was a lack of knowledge on the removal of MPs in SWTPs. A  
 704 SWTP with two treatment routes was sampled for this study. Figure 3 showed the distribution of  
 705 MPs in quantity and types, together with the removal/retention efficiency of MPs by different  
 706 stages of the treatment. The up-bottom order of water types marked on the Y-axis was consistent  
 707 with the discharge sequence in SWTP, and the samples was named with the related treatment  
 708 process (the former or the latter). Notably, the rules for making figures were kept for all samples  
 709 taken from different water treatment plants in this study. In addition, the evolution of MPs in  
 710 surface area, and in major and minor dimensions of samples was shown in Figure 4 and Figure S1,  
 711 respectively.

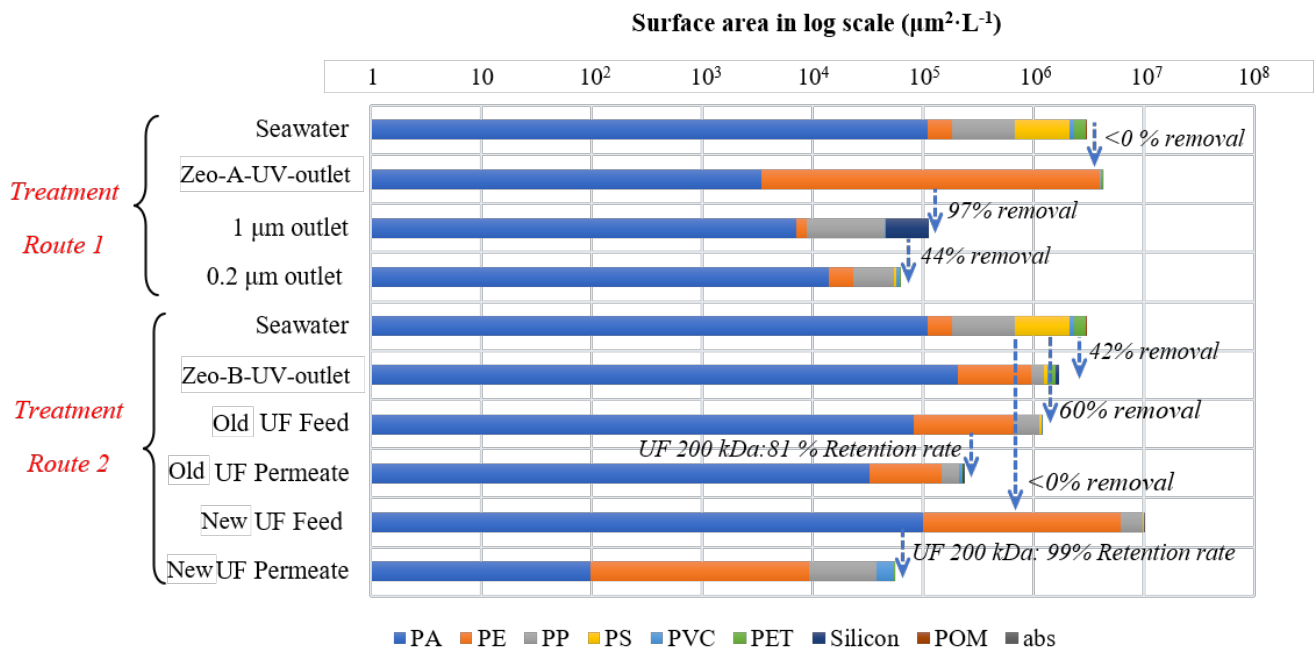


712

713 Figure 3 Concentration in quantity and types of MPs in seawater samples together with the related  
 714 retention/removal rates by different treatment processes (retention used for membranes, removal used for  
 715 the other treatments)

716

717



718

719 Figure 4 Concentration in surface area and types of MPs in seawater samples together with the related  
720 retention/removal rates by different treatment processes (retention used for membranes, removal used for the  
721 other treatments)

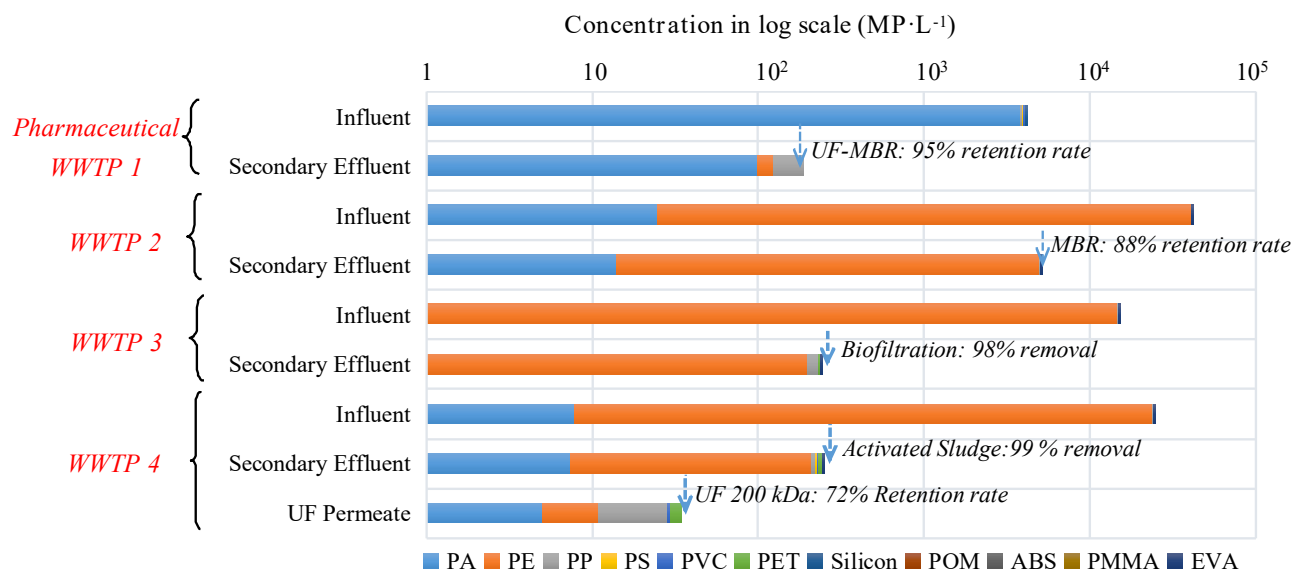
722 In treatment route 1 and in view of quantity (Figure 3), the entire treatment process (from  
723 seawater to 0.2  $\mu\text{m}$  filter outlet) provided  $94 \pm 2\%$  total removal rate of MPs. Zeo-A-UV (Zeolite  
724 A and UV) process removed  $63 \pm 4\%$  of MPs from seawater, followed by  $83 \pm 4\%$  removal by 1  
725  $\mu\text{m}$  cartridge, while almost no MPs in quantity was removed by 0.2  $\mu\text{m}$  cartridge from 1  $\mu\text{m}$  outlet.  
726 In view of surface area (Figure 4), Zeo-A-UV process showed no removal but increased MPs in  
727 Zeo-A outlet, while 1  $\mu\text{m}$  cartridge and 0.2  $\mu\text{m}$  cartridge contributed to  $97 \pm 8\%$  and  $44 \pm 28\%$   
728 removal of MPs, respectively. The different results of MPs in quantity and surface area reflected  
729 the limits of expressing the results in  $\text{MPs}\cdot\text{L}^{-1}$  sole for MPs description, and the supplement of the  
730 surface or dimensions of MPs was necessary for more accurate descriptions. In treatment route 2:  
731 Zeo-B-UV outlet, Old UF feed, and New UF feed provided increased MPs concentration in  
732 quantity compared to seawater (Figure 3), yet resulted in  $42 \pm 5\%$ ,  $60 \pm 11\%$ ,  $<0\%$  removal of  
733 MPs in surface area (Figure 4), respectively. While Zeo-B-UV outlet, Old UF feed, and New UF  
734 feed were samples all treated by primary sedimentation + 100  $\mu\text{m}$  disk + Zeolite B process, and  
735 Zeo-B-UV outlet, the increased MPs in quantity reflected the external MPs contaminations (mainly  
736 by PA, PE, PP, and PS) in sampling sites or transportation channels after partial removal of MPs  
737 by treatment process. However, the followed UF membranes, especially the New UF, exhibited  
738 excellent retention ability on MPs both in quantity and surface area whatever the MPs in UF feeds.  
739 The Old UF and New UF, respectively, supported  $74 \pm 1\%$  and  $96 \pm 2\%$  retention of MPs in  
740 quantity, and supported  $81 \pm 9\%$  and  $99.5 \pm 0.5\%$  retention of MPs in surface area. Around 130  
741  $\text{MP}\cdot\text{L}^{-1}$  was detected in Old UF permeate, and less than 21  $\text{MP}\cdot\text{L}^{-1}$  were obtained in New UF  
742 permeate. Therefore, the results indicated that UF membranes provide effective retention on MPs  
743 from influent, especially with recent UF membranes.

744 In general, SWTP treatments by 1  $\mu\text{m}$  cartridge, 0.2  $\mu\text{m}$  cartridge, and UF membranes all  
745 provided comparatively considerable removal or retention efficiency on MPs, while treatments  
746 with Zeolite A (+UV) and Zeolite B (+UV) posed potential contaminations of MPs. When treated  
747 with Zeolite A (+UV) (Figure 3 and Figure 4), 96.4% of PE particles in total surface area was  
748 invaded in Zeo-A-UV outlet, while only 2.4% of PE occurred in seawater (raw influent). Moreover,

749 Zeo-A-UV outlet was found with more and larger MPs with  $d_{\min} > 150 \mu\text{m}$  and  $d_{\text{maj}} > 400 \mu\text{m}$   
750 compared to that in seawater (SI Figure S2(a)(b)). When treated with Zeolite B (+UV), increasing  
751 of PA, PE, and PP particles in quantity were discovered in Zeo-B-UV outlet compared to seawater.  
752 Since zeolites were the aluminosilicate members of microporous solids, mainly consisting of  
753 silicon, aluminum, and oxygen (Jiang et al., 2018), micro-pollution including MPs could be  
754 blocked in zeolite in long-term use and without cleaning, which could lead to secondary pollution  
755 of MPs in the effluents. This result reminds to pay attention to the secondary contamination of  
756 microplastics which might come from the treatment processes and pipes. In view of UF membranes,  
757 both Old and New UF showed good retention rates on MPs in quantity and in surface area.  
758 According to SI Figure S2, more smaller MPs within  $d_{\min} < 40 \mu\text{m}$  and  $d_{\text{maj}} < 50 \mu\text{m}$ , occupying  
759 65% of total MPs, were discovered in Old UF permeate (SI Figure S2 (g)). Meanwhile, MPs with  
760  $d_{\min} > 80 \mu\text{m}$  and  $d_{\text{maj}} > 200 \mu\text{m}$  were almost completely retained by both Old UF and New UF  
761 membranes. Since the two UF modules were both 200 kDa hollow fiber membrane made of PES,  
762 the main difference was the duration of use. It could be inferred that UF membranes themselves  
763 were not the polluting sources of MPs according to the MPs types in permeates, while membrane  
764 aging could decrease the MPs retention capacity especially on smaller particles ( $d_{\min} < 40 \mu\text{m}$  and  
765  $d_{\text{maj}} < 50 \mu\text{m}$ ). Additionally, comparing the removal/retention rates of MPs in quantity (Figure 3)  
766 and surface area (Figure 4) by different treatment processes (e.g. 1  $\mu\text{m}$  cartridge, 0.2  $\mu\text{m}$  cartridge,  
767 and UF membranes without ignorable external contaminations), higher removal rate in total  
768 surface area was presented compared to that in quantity. This study is one of the first to report this  
769 finding and it needs more verifications in the followed investigations and further discussion for  
770 explanations.

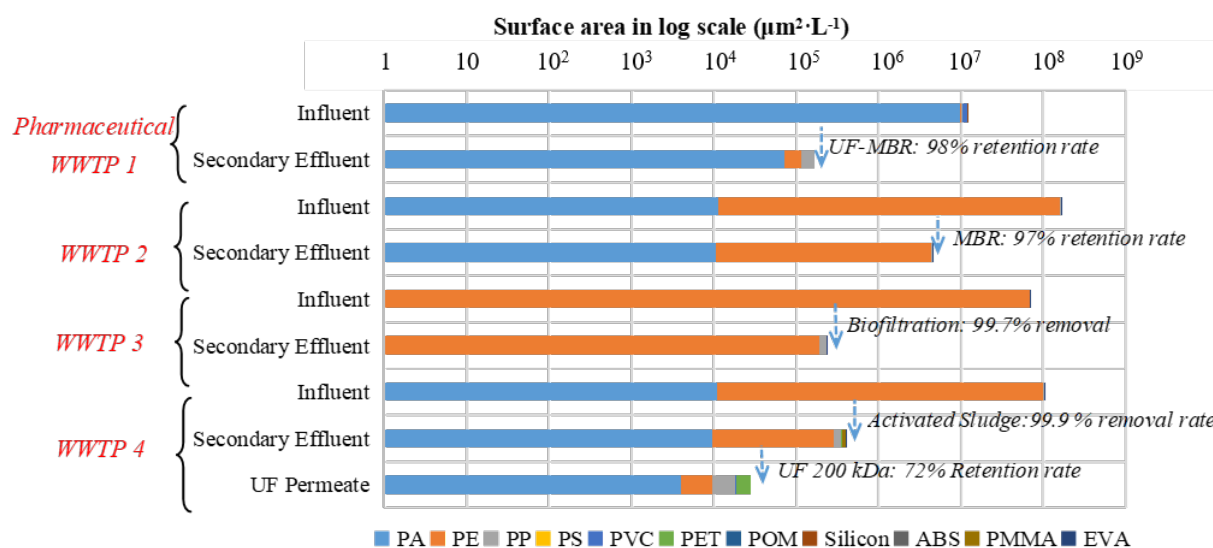
### 771 3.3. Microplastics Retention in Wastewater Treatment Plants

772 Four different WWTPs in France were sampled for MPs analysis. Three of WWTPs (WWTP  
773 1-3) built with secondary treatments, and WWTP 4 was equipped upgraded with UF as a tertiary  
774 treatment. The distribution and removal/retention of MPs in quantity and surface area were shown  
775 in **Erreur ! Source du renvoi introuvable.** and **Erreur ! Source du renvoi introuvable.**,  
776 respectively. The distribution of major/minor dimensions of MPs in different WWTPs was shown  
777 in Figure S3.



778

779 Figure 5 Concentration in quantity and types of MPs in WWTPs samples together with the related  
 780 retention/removal rates by different treatment processes (retention used for membranes, removal used for  
 781 other treatments)



782

783 Figure 6 Concentration in surface area and types of MPs in WWTPs samples together with the related  
 784 retention/removal rates by different treatment processes (retention used for membranes, removal used for  
 785 other treatments)

786 In terms of MP types, PE was the main type of MPs in municipal WWTPs (WWTP 2–4), and  
 787 PA was predominant in the pharmaceutical WWTP 1 (**Erreur ! Source du renvoi introuvable.**

788 and **Erreur ! Source du renvoi introuvable.**). In terms of concentration in quantity (**Erreur !**  
789 **Source du renvoi introuvable.**), the influent of pharmaceutical WWTP 1 contained around 4,203  
790  $\text{MP}\cdot\text{L}^{-1}$  of MPs, while those of municipal WWTPs were more loaded with MPs ranging from  
791 15,000  $\text{MP}\cdot\text{L}^{-1}$  to 42,000  $\text{MP}\cdot\text{L}^{-1}$ . In municipal WWTPs, the removal rates of MPs by activated  
792 sludge, physico-chemical and biofiltration, and MBR all achieved up to >88% in quantity (**Erreur !**  
793 **Source du renvoi introuvable.**), together with > 97% removal of MPs in surface area (**Erreur !**  
794 **Source du renvoi introuvable.**). Focusing on membrane processes, both MBRs in WWTP 1 and  
795 WWTP 2 showed good retention rates (88%-95% in quantity and 97%-98% in surface area) on  
796 MPs. As MBR permeates in WWTP 1 and WWTP 2 respectively contained 190  $\text{MP}\cdot\text{L}^{-1}$  and 5,065  
797  $\text{MP}\cdot\text{L}^{-1}$ , the MBR in WWTP 1 (pore size of 0.02  $\mu\text{m}$ ) was more effective on MPs removal. By  
798 comparison, MBR in WWTP 2 showed weaker retention of MPs than in WWTP 1 which might  
799 be caused by bigger pore size of membrane in WWTP 2 (0.1  $\mu\text{m}$  > 0.02  $\mu\text{m}$ ) but also by 10 times  
800 higher MPs concentration (both in quantity and surface area) in WWTP 2 influent. Although the  
801 retention rate by 200 kDa UF in WWTP 4, 72% in quantity and 72% in surface area, was lower  
802 than secondary treatments in all WWTPs (>88%), UF membranes enabled the lowest MPs  
803 concentration both in quantity (35  $\text{MP}\cdot\text{L}^{-1}$ ) and surface area. The relative lower retention rates by  
804 UF were mainly due to the reduced MPs concentrations in UF permeate. Finally, even the  
805 concentration of MPs varied greatly in raw wastewater, all secondary treatments provided  
806 considerable removal efficiency on MPs both in quantity and surface area. However, although the  
807 MPs removal by secondary treatment was quite high, the residue MPs in secondary effluents  
808 ranging from 128  $\text{MP}\cdot\text{L}^{-1}$  to 5,065  $\text{MP}\cdot\text{L}^{-1}$  still needs more treatment. Thereby, UF membrane was  
809 recommended as the effective treatment for MPs after secondary treatment to better control the  
810 MPs concentrations (quantity and surface area) at lower levels.

811 In view of dimensions (Figure S3), the dimensions of MPs in MBR effluent and UF permeate  
812 were smaller than that in activated sludge or biofiltration process effluent. Particularly, 200 kDa  
813 UF in WWTP 4 controlled MPs within a much smaller range,  $d_{\min} < 80 \mu\text{m}$  and  $d_{\text{maj}} < 200 \mu\text{m}$ .  
814 Additionally, the removal rates of MPs by each treatment in surface area was also higher than that  
815 in quantity in all WWTPs, as for SWTP samples. Considering the dimensions of MPs that can be  
816 removed, smaller particles, especially those with  $d_{\min} < 40 \mu\text{m}$  and  $d_{\text{maj}} < 50 \mu\text{m}$  increased the  
817 difficulty for MPs removal/retention (in quantity). In agreement with previous studies on the  
818 effectiveness of membrane processes for controlling MPs (Padervand et al., 2020; Talvitie et al.,

819 2017), MBR and UF process in this study both showed considerable retention rates of MPs and  
820 significant decrease of MPs in dimensions and quantity, and UF membrane is suggested for  
821 terminal water treatment due to the lowest MPs concentrations in UF permeate.

#### 822 3.4. Microplastics Retention in Drinking Water Treatment Plants

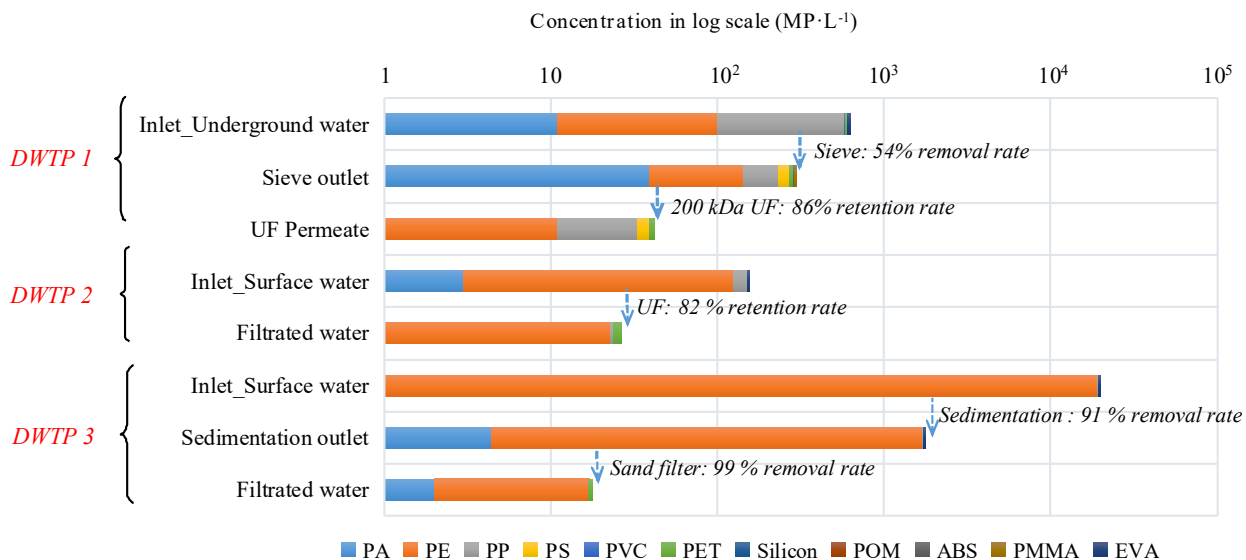
823 In DWTPs, the distribution and removal/retention of MPs in quantity and surface area were  
824 shown in Figure 7 and Figure 8, respectively. The distribution of major/minor dimensions of MPs  
825 in different DWTPs was shown in Figure S4.

826 In terms of MPs types in quantity (Figure 7), PE was the most dominant in surface water  
827 feeding DWTP 2 and DWTP 3, occupying  $80 \pm 3\%$  and  $98 \pm 0.2\%$  in inlets, respectively. PP,  
828 occupying  $76 \pm 6\%$ , was the main component in underground water based DWTP 1. However,  
829 considering about the surface area (Figure 8), the proportion of PE in inlet of DWTP 3 was  
830 decreased to  $56 \pm 1\%$ , while PS was secondly dominant in inlet of DWTP 3 occupying  $44 \pm 2\%$   
831 in total surface area, while it occupied only  $0.5 \pm 0.1\%$  in quantity meaning that PS particles have  
832 much bigger dimensions than PE particles. PS particles in inlet of DWTP 3 were detected in fewer  
833 counts but in larger surface area (Figure 7 and Figure 8). The results demonstrated the importance  
834 of three-item characterization of MPs (counts, surface area, and types of MPs) in this study. In  
835 view of the concentrations, the distribution of MPs differed a lot in inlets of different DWTPs,  
836 ranging from the lowest concentration of  $153 \text{ MP} \cdot \text{L}^{-1}$  in DWTP 2 to the highest concentration of  
837  $19,836 \text{ MP} \cdot \text{L}^{-1}$  in DWTP 3, averagely in quantity.

838 Focused on treatment process, the primary treatment by sieve in DWTP 1 enabled  $54 \pm 3\%$   
839 and  $54 \pm 1\%$  removal of MPs in quantity and surface area, respectively. The primary treatment by  
840 sedimentation in DWTP 3 provided higher  $91 \pm 4\%$  and  $95 \pm 2\%$  removal of MPs in quantity and  
841 surface area, respectively. Treatment of filtration in all DWTPs were positioned in the  
842 terminal/tertiary treatment, resulting in qualified permeates with lowest concentration of MPs in  
843 each DWTP. The sand filter in DWTP 3 retained  $99 \pm 1\%$  of the MPs in quantity, and  $99 \pm 1\%$  in  
844 surface area. Additionally, the UF membranes both in DWTP 1 and DWTP 2 showed 80-88 %  
845 retention of MPs in quantity, and 90-95% retention of MPs in surface area. Additionally, the MPs  
846 concentration in filtration permeates from all DWTPs was effectively reduced to 18-42  $\text{MP} \cdot \text{L}^{-1}$   
847 whatever the inlet water conditions. In view of MPs dimensions (Figure S4), UF membranes as

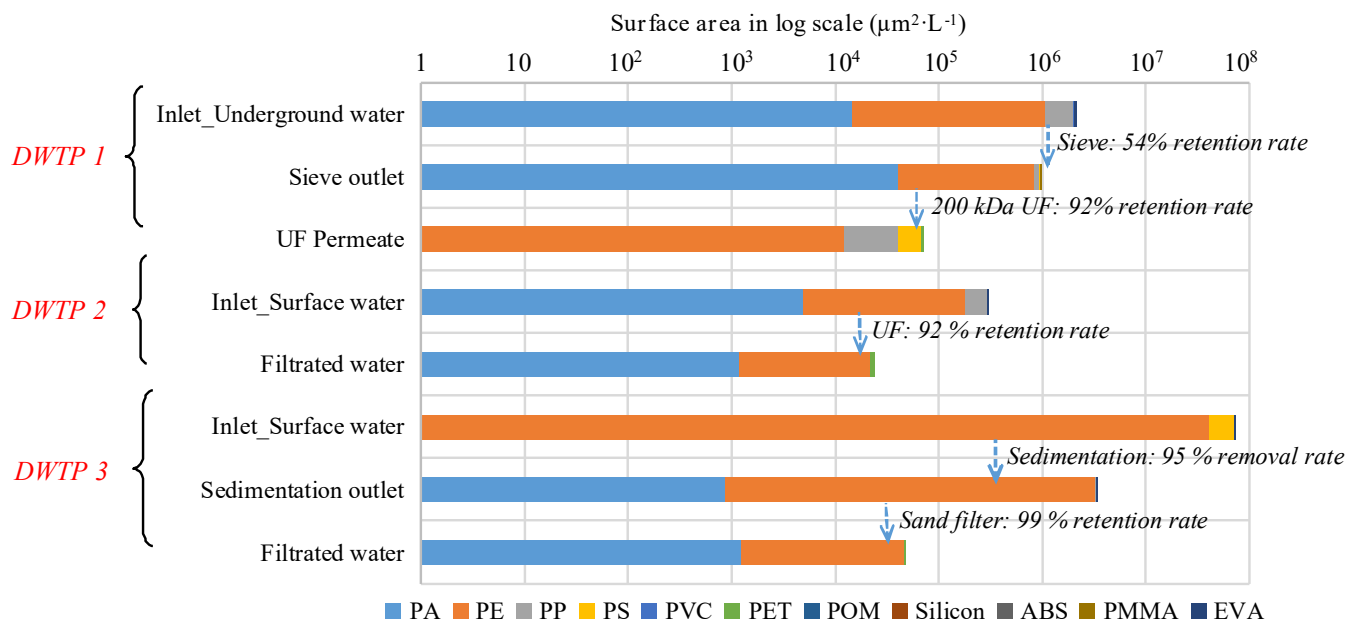


848 well limited MPs dimensions within  $d_{min} < 80 \mu m$  and  $d_{maj} < 200 \mu m$ . UF membranes were proved  
 849 to be effective for MPs retention from DWTPs. The removal/retention rates of MPs by each  
 850 treatment process in surface area was also higher than the one in quantity in DWTPs.



851

852 Figure 7 Concentration in quantity and types of MPs in DWTPs samples together with the related  
 853 retention/removal rates by different treatment processes (retention used for membranes, removal used for  
 854 the other treatments)



855

856

857 Figure 8 Concentration in surface area and types of MPs in DWTPs samples together with the related  
858 retention/removal rates by different treatment processes (retention used for membranes, removal used for  
859 the other treatments)

## 860 4. Discussion and Conclusion

861 In the light of above, most of the considered treatment processes are effective on MPs  
862 removal/retention, except some process with external pollution or the secondary release of MPs.  
863 Among all treatment processes, UF membranes, 0.2-1  $\mu\text{m}$  cartridges, and sand filter as the terminal  
864 treatment enablede considerable retention rates on MPs to ensure the good quality of final effluent.  
865 Particularly, UF membrane is highly suggested for MPs retention whatever the water types. UF  
866 membranes could provide acceptable MPs concentrations in permeates and almost complete  
867 retention rate on MPs of  $d_{\text{min}} > 80 \mu\text{m}$  and  $d_{\text{maj}} > 200 \mu\text{m}$ . The retention rates of 200 kDa UF in  
868 SWTP, WWTPs, and DWTPs ranged from 72-98% in quantity to 81-99% in surface area. However,  
869 considering that most transportation pipes and connections were made of plastics as well as the  
870 dimensions of MPs could be smaller to nano sizes, it was hard to say that a certain process can  
871 remove MPs completely, even with UF membranes. The full mechanisms of MPs transfer in the  
872 processes still need to be furtherly studied in the future.

873 Table 2 summarized the performances of all treatment processes on MPs dimension ranges as  
874 well as concentration and proportion of smaller MPs (Min-d  $< 40 \mu\text{m}$  Maj-D  $< 50 \mu\text{m}$ ) in this study.  
875 The results revealed the effectiveness on MPs removal/retention might ultimately depend on the  
876 pore size of treatment processes without chemical reactions. By statistics, the counts of smaller  
877 MPs with  $d_{\text{min}} < 40 \mu\text{m}$  and  $d_{\text{maj}} < 50 \mu\text{m}$  were ranked in dominant positions, especially in secondary  
878 and tertiary effluents, accounting for 11-74% of MPs in quantity. However, the presence of smaller  
879 MPs increased the difficulty for detection, tracking, and removal, and increased the potential threat  
880 for natural environment and organisms. According to Batel et al. (2020), cellular uptake and  
881 transfer of MPs across organs seemed only possible for smaller microsized or nanosized plastic  
882 particles. Besides, the translocation of smaller MPs (0.1-150  $\mu\text{m}$ ) into the tissue has been  
883 demonstrated in multiple studies conducted with rodents, rabbits, dogs, and humans (Carr et al.,  
884 2012; Imhof and Laforsch, 2016; Larue et al., 2021). Due to higher surface-to-volume ratios,  
885 smaller MPs might deliver higher amounts of contaminants such as persistent organic pollutants

886 through transport in gut system and accumulation in food chains (Batel et al., 2020). Therefore, it  
 887 revealed that the identification, quantification, and removal of smaller MPs or even nano-sized  
 888 plastics would be a challenge for future studies.

889 Table 2 The comparison of different treatment processes on microplastics removal in this study

Plant	Main Process	Related effluent	Minor- Dimension ( $\mu\text{m}$ )	Major- Dimension ( $\mu\text{m}$ )	MP (Min-d <40 $\mu\text{m}$ Maj-D <50 $\mu\text{m}$ )	
					Concentration ( $\text{MP}\cdot\text{L}^{-1}$ )	Proportion
SWTP	Zeolite + UV	Zeo-A-UV/Zeo-B-UV outlet	<500	>600	16–194	10–38%
	1 $\mu\text{m}$ cartridge	1 $\mu\text{m}$ outlet	<150	<400	~3	~11%
	0.2 $\mu\text{m}$ cartridge	0.2 $\mu\text{m}$ outlet	<150	<400	~10	~37%
	Old UF (200kDa)	Old UF permeate	<150	<600	~80	~62%
	New UF (200kDa)	New UF permeate	<80	<400	~11	~52%
WWTP 1	UF-MBR (0.02 $\mu\text{m}$ )	Secondary effluent	<80	<200	~135	~71%
WWTP 2	MBR	Secondary effluent	<150	<400	~3585	~71%
WWTP 3	Physico-chemical with biofiltration	Secondary effluent	<150	<400	~180	~74%
WWTP 4	Activated sludge	Secondary effluent	<250	<400	~122	~49%
	200 kDa UF	UF permeate	<80	<200	~22	~63%
DWTP 1	Sieves	Sieve outlet	<250	>600	~173	~59%
	200 kDa UF	UF permeate	<80	<200	~22	~52%
DWTP 2	UF	Filtrated water	<80	<200	~19	~70%
DWTP 3	Sedimentation	Sedimentation outlet	<250	>600	~1098	~62%
	Sand filter	Filtrated water	<250	>600	~6	~33%

890 This study evidenced the ubiquitous microplastics in seawater, wastewater, surface water, and  
 891 groundwater of France. In comparison, wastewater was observed to aggregate the most MPs  
 892 compared to seawater, surface water, and ground water, due to urban agglomeration and  
 893 anthropogenic impact. The 4203-42,000  $\text{MP}\cdot\text{L}^{-1}$  of MPs in raw wastewater of France by  $\mu$ -FTIR

894 in this study was positioned at the highest level among European countries (57.6-10,044 MP·L<sup>-1</sup>),  
895 USA (1-133 MP·L<sup>-1</sup>), and Asia (<500 MP·L<sup>-1</sup>), etc (Long et al., 2019; Sun et al., 2019; Wu et al.,  
896 2022). The results in France was much closer to MPs abundance in Denmark which ranked just  
897 after France with 2,223-10,044MP·L<sup>-1</sup> in WWTPs (Simon et al., 2018). In terms of surface water  
898 and groundwater, the detected MPs in inlet of DWTPs in France (153-19,836 MP·L<sup>-1</sup>) were as well  
899 the highest levels among most other countries and regions based on the survey by Selvam et al.  
900 (2021) with 1-20 MP·L<sup>-1</sup> and by Li et al.(2018) with <187 MP·L<sup>-1</sup>. The MPs abundances in DWTP  
901 1-2 (153-632 MP·L<sup>-1</sup>) was in the same order of magnitude than some studies on rivers, such as the  
902 Lek Canal with 460 MP·L<sup>-1</sup> (Bäuerlein et al., 2022) and Amsterdam canals with 48–187 MP·L<sup>-1</sup>  
903 (Leslie et al., 2017). In terms of seawater, MPs abundance in seawater of France was also  
904 positioned at a high level compared to others, such as in Asia and Europe, Pacific, Southern, and  
905 Atlantic Oceans (Zhang et al., 2022; Zheng et al., 2019). Additionally, the four WWTPs, three  
906 DWTPs, and SWTP in France provided by 87.8-99.8%, 82.3-99.9%, 69.0-96.0% removal of MPs  
907 in quantity and showed comparative effectiveness to water treatment plants across worldwide  
908 countries (Wu et al., 2022) . The three DWTPs in France provided significant removal of MPs  
909 from the inlet. Moreover, the types of MPs in water bodies of France are basically the same as  
910 those in other countries or regions, mainly composed of PE, PP, PS, and PA (Long et al., 2019;  
911 Mintenig et al., 2020; Wang et al., 2020). However, as most studies did not show the surface area  
912 of MPs in water bodies, the comparison on MPs concentration and removal/retention capacity  
913 based on surface area was not discussed in this study. In summary, quantitative concentration of  
914 MPs in different water bodies of France in this study were all assessed at high levels, partially  
915 because of the higher resolution of the detection method (Part I). Indeed, one should keep in mind  
916 that the results found in the literature may unfortunately still be very method-dependent (collection,  
917 detection, analytical method...). but still it reminds to researchers and public that they should pay  
918 more attention on MPs management and treatment.

919 In conclusion, this study used a quantitative and qualitative analytical methodology of MPs by  
920  $\mu$ -FTIR spectroscopy carried with the siMPle analytical software which has been confirmed to be  
921 an efficient, reliable, and non-destructive detection method in Part I. For the first time, with the  
922 same analytical method, MPs were analyzed in different water types from France and different  
923 treatment processes were assessed for their MP removal, in a single paper. The MPs concentration  
924 in different types of water resources varied a lot. Generally, raw wastewaters contained the highest

925 quantity of MPs, then generally followed by some surface waters, groundwater. PE was the most  
926 abundant type of MPs in almost all water types, then followed by PP, PS, and PET. Most treatment  
927 technologies, including primary, secondary, and tertiary treatment had the ability to remove/retain  
928 MPs from feed water. The efficiency of treatment processes including ultrafiltration should not  
929 make us forget that the retained MPs must also be managed efficiently. Some treatment processes,  
930 transport pipes, or other external instruments could also cause a secondary MPs. Moreover, almost  
931 in all treatment process, the removal/retention rates on MPs in surface area were higher than that  
932 in quantity. The possible reasons included (1) feed water contained more larger pieces of MPs  
933 which were preferably removed/retained by different processes; (2) the attachment of smaller MPs  
934 on larger MPs was separated under forces of water flow and interception; (3) aged MPs in natural  
935 waters were prone to undergo physical abrasion, biofouling, and chemical oxidation-associated  
936 weathering by different treatments and further decomposed into smaller secondary MPs (Wu et al.,  
937 2022). It is believed that this study could finally improve public awareness on MPs contamination,  
938 help MPs limitation policies, and select appropriate removal technology in the future.  
939

## 940 5. Acknowledgment

941 The authors would like to warmly thank Alexandre Michelet and Jean-Philippe Mélis from  
942 PerkinElmer for provisioning them with the FTIR microscope and for helping to use it, Stéphanie  
943 Lebarillier (LCE lab) for the technical support, the ECCOREV Research Federation (FR3098) and  
944 and ITEM Research and Teaching Institute from Aix-Marseille University for their financial  
945 support.

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