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Screening of microplastics in water and sludge lines of a drinking water treatment plant in Catalonia, Spain

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ABSTRACT

Microplastics (MPs) are emerging pollutants detected everywhere in the environment, with the potential to harm living organisms. The present study investigated the concentration, morphology, and composition of MPs, between 20 μm and 5 mm, in a drinking water treatment plant (DWTP) located close to Barcelona (Catalonia, NE Spain). The sampling included different units of the DWTP, from influent to effluent as well as sludge line. Sampling strategy, filtration, allows sampling of large volumes of water avoiding sample contamination, and during 8 h in order to increase the representativeness of MPs collected. The pre-treatment of the samples consisted of advanced oxidation with Fenton's reagent and hydrogen peroxide, followed by density separation of the particles with zinc chloride solution. Visual identification was performed with an optical and stereoscopic microscope with final Fourier-transform infrared spectroscopic (FTIR) confirmation. MPs were found in all DWTP samples, with concentrations from 4.23 ± 1.26 MPs/L to 0.075 ± 0.019 MPs/L in the influent and effluent of the plant, respectively. The overall removal efficiency of the plant was 98.3%. The most dominant morphology was fibers followed by fragments and films. Twenty-two different polymer types were identified and synthetic cellulose, polyester, polyamide, polypropylene, polyethylene, polyurethane, and polyacrylonitrile were the most common. Although MPs could be incorporated from the distribution network, MPs intake from drinking water from this DWTP was not an important route compared to fish and seafood ingestion.

1. Introduction

Microplastics (MPs) are defined as an emergent pollutant that is ubiquitously detected in the natural environment (marine and freshwater, soil and air) (Franco et al., 2021) posing a threat to the health of living organisms (Alimi et al., 2018; Chen et al., 2020; Waring et al., 2018). MPs are synthetic, non-biodegradable polymers with a diameter size of less than 5 mm. Divided by origin, they can be either primary (directly manufactured microbeads and fibers), or secondary (breakdown of larger plastic). Primary MPs come mainly from personal care products and textile fibers, while secondary ones are from weathering, photolysis, and decomposition of discarded plastics in the environment (Expósito et al., 2021; Ziajahromi et al., 2017). MPs may act as vectors for environmental pollutants, hazardous chemical additives, and pathogen microorganisms which have the potential to be spread through the food chain (Prata, 2018; Rist et al., 2018; Rodrigues et al., 2019).

Food and water intake have been identified as a route of exposure to MPs. They have been detected in numerous samples of commercially available seafood, bottled water, tap water, honey, and salt (Iñiguez et al., 2017; Liebezeit and Liebezeit, 2015; Mason et al., 2018; Mintenig et al., 2019; Pivokonsky et al., 2018; Tanaka and Takada, 2016; Yang et al., 2015). To date, toxicological studies on the possible effects of MPs on human health have not been fully established (Revel et al., 2018; WHO, 2019; Wright and Kelly, 2017) mainly due to challenges in the

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experimental design and determining the actual MPs concentration to which organisms are exposed. However, a recent study found that MPs could alter human microbiota (Tamargo et al., 2022). It is worth noting that, in addition to ingestion, inhalation of MPs present in the air, as well as dermal contact, are also routes of exposure that must be considered (Prata et al., 2020).

MPs pollution receiving considerable attention is also evidenced by the number of publications that have drastically increased from 2011 until now (Sol et al., 2020). Additionally, MPs have been mentioned in the European drinking water Directive (2020) to be included in the risk assessment of the DWTP and their supply system. However, there is a lack of published data addressing the problem of MPs contamination in drinking water treatment plants (DWTP) (Wang et al., 2020). Drinking water treatment plants ensure the quality and safety of drinking water, thus it is urgent to investigate the fate of MPs in different treatment processes. Currently, there are no legislative restrictions on the abundance of MPs in drinking water (Novotna et al., 2019; Wang et al., 2020). Only a few studies investigated MPs based on full-scale DWTP. Pivokonský et al. (2020) investigated the occurrence and fate of microplastic particles, up to 1 μm, at two different DWTPs (with advanced and conventional treatment systems), within one river catchment in the Czech Republic, and discovered a significant difference between them: DWTP containing flocculation, sedimentation, sand filtration, and granular activated carbon filtration units had MPs removal efficiency higher than 80%, unlike the other DWTP with flocculation and sand filtration units, that had MPs removal rate of only 40%. Other studies by Wang et al. (2020) and Shen et al. (2021) also reported a high removal rate of DWTPs located in China, both greater than 85%. Furthermore, characterization of MPs in eight DWTPs located in the UK showed that is possible to eliminate over 99.99% of MPs from their source water (river, groundwater, or an upland reservoir) leading to its accumulation in dry sludge (Johnson et al., 2020). Although MP's removal rates in DWTP are high, several MPs are still found in the effluents of DWTP. Investigating MPs in a size range of 20 μm to 5000 μm, Mintenig et al. (2019) reported that the concentration of MPs in the German DWTP effluent was 0.007 MPs/L while Dalmau-Soler et al. (2021) reported the concentration of 0.06 ± 0.04 MPs/L, in the effluent of Spanish DWTP, for the same size range. This study performed by Dalmau-Soler et al., (2021) investigated the presence of microplastics along the same river basin as in the present study, however different drinking water treatment plant was observed. The microplastic methodology is similar in both studies allowing better comparison of the results such as identification method (FTIR spectroscopy), analyzed size (20μm to 5 mm), and units used to report the MPs levels (MPs/L). Furthermore, although the plant is different, the treatments in both cases are similar where the plant from the previous study consists of clarifiers, sand filtration, ozonation, GAC filtration and ultrafiltration, and reverse osmosis. Some studies have also found that during the water treatment in the plant, the number of MPs increases, such as a recent study that reported the reduction of overall removal from 70 to 52% in a Clearwell (Cherniak et al., 2022). Information about the presents of MPs in the effluents of DWTP cannot be ignored, since the treated water is intended for the local population.

The aim of the present study is to investigate the concentrations, morphology, and composition of MPs in different treatment units of a river freshwater supplied DWTP including both, water and sludge lines. Removal efficiency was calculated to determine which treatment unit had the greatest impact on MPs removal and finally, to estimate the intake of MPs by drinking water from this source.

2. Materials and methods

2.1. Drinking water treatment plant characteristics

The studied DWTP of Llobregat river is located near Barcelona (NE Spain) and has a treatment capacity of 3.2 m^3/s. The DWTP consists of the treatment units depicted in Fig. 1. Briefly, the first treatment is water roughing in which the surface river water (pH 8) passes through bars that prevent large materials, suspended in the water, from entering the plant. The water then flows through three grit channels. Potassium permanganate is added (0.5–1.5 mg/L) for the pre-oxidation of organic and inorganic compounds. In the next step, the water enters the mixing chambers, where pH is adjusted (7.4–7.7) with carbon dioxide, and chemicals such as coagulants and flocculants (Aluminium Polychloride 25 mg/L and PolyDADMAC 0.8 mg/L) were added for turbidity removal. The water then goes to eight clarifiers (Pulsator®), where the flocs present in the water are sedimented and the excess sludge is discharged. After clarification, oxidation with chloride dioxide is performed. The next process is filtration in two steps in twelve sand filters (70 cm thickness). Three of them are filled with two layers of sand (0.8–1.6 and 1.4–2.5 mm grain size) and the other nine are filled with one layer (1–2 mm grain size) together with sand Filtralite® material. This step is used not only for physical filtration but also for biofiltration. Sand filtration processes were followed by a granular activated carbon (GAC) filtration process, 15 filters of 150 cm thickness, Norit GAC1240, where part of the dissolved organic matter is adsorbed (organochlorides). Part of the treated water with the conventional treatment undergoes microfiltration with 10 μm polypropylene cartridge filters and reversible electrodialysis (EDR), which removes a high percentage of salt content in treated water. Then water is remineralized with calcium hydroxide in solution and carbon dioxide injection. After that, the remineralized water is mixed with the water non-treated with microfiltration, and EDR units. Final disinfection is done with a sodium hypochlorite solution, and water is stored in two tanks of 213,000 m^3 before the distribution.

2.2. Sampling methodology

Microplastic samples from the 6 water line sampling points were obtained by filtration from DWTP in May 2021 for 8 h. A total of 360L (3 replicates of 120L each) of river water (influent) from the field inside the plant for 8 h were filtered through a set of stainless-steel sieves (Cisa Sieving Technologies, 200 mm diameter x 50 mm height) with mesh size: 20, 50, 100, 500 and 1000 μm. To avoid contamination, the other water line samples were taken from the taps of the DWTP laboratory, from the effluents of treatment units (clarifier lines 1 ($n = 7$) and 2 ($n = 7$), sand filter ($n = 3$), and activated carbon filter unit ($n = 3$)) and the effluent of the plant ($n = 7$). Water was filtered through an in-line stainless filter holder containing a PTFE filter (*Sartorius*, 10 μm pore size) connected to a tap for 8 h. The total volumes of water sampling were 29L, 28L, 267L, 600L, and 965L for clarifier line 1 effluent, clarifier line 2 effluent, sand filter effluent, carbon filter effluent, and effluent of the plant, respectively. Water volume filtered varied from one treatment unit's effluent to another according to the expected MPs levels (Mintenig et al., 2019; Pivokonsky et al., 2018). Stainless steel sieves covered in aluminum foil and PTFE filters stored in glass Petri dishes were immediately transferred to the laboratory for further analysis. This sampling approach allows us to sample relatively large volumes of water, near 1m^3 in the effluent of the plant, avoiding sample contamination and during large periods (8 h). More information regarding sampled volume and the replicates are summarized in the supplementary data. Finally, dry sludge (300n g) ($n = 1$) and water from centrifugation process (5 L) ($n = 1$) were taken from the operation unit of the sludge line, in glass bottles pre-cleaned with filtered water. Glass bottles were covered with aluminum foil to prevent contamination from the cap.

2.3. Analysis of samples in the laboratory

In order to perform pre-treatment of the samples, the following chemicals and materials were used: absolute ethanol (*Scharlau*, >99.9%), iron (II) sulfate heptahydrate (*Sigma-Aldrich*, >99.0%), hydrogen peroxide (*PamReac AppliChem ITW Reagents*, 30%), PTFE-filter (*Sartorius*, 5 and 10 μm pore size), and zinc chloride (*Acros*

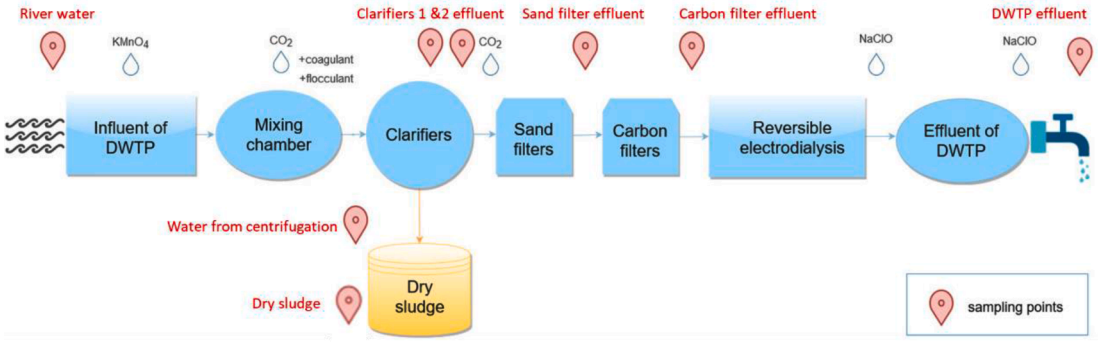

Fig. 1. Flow diagram of the drinking water treatment plant (DWTP) units and sampling points.

Organics, >98.0%).

Nowadays there are no standard procedures for measuring MPs in drinking water treatment plants (Conesa and Ortuño et al., 2022). Pre-treatment of the samples was performed in order to extract MPs and remove all organic and inorganic matter to avoid interference. Three versions of the same protocol are established in the laboratory based on the requirements and nature of the samples (from highest organic matter content in dry sludge or centrifugated water to lowest organic matter content in water from clarifiers, sand/carbon filters, or effluent). In general, the protocol consists of oxidation to remove organic matter. Oxidation with Fenton's reagent was recommended by US National Oceanic and Atmospheric Administration (NOAA) for MPs analysis in marine water (Masura et al., 2015) and other authors have also used it as a purifying step in microplastic identification from different water samples, sediments and organisms (Erni-Cassola et al., 2017; Karami et al., 2016). Hydrogen peroxide is another oxidizing agent that can digest organic matter with little or no polymer degradation (Nuelle et al., 2014; Qiu et al., 2016; Zhao et al., 2017). In order to remove inorganic matter from the samples, density separation was applied using a salt solution (Ziajahromi et al., 2017). In the present work, zinc chloride solution (density: 1.8 g/mL) was chosen because its density is suitable for successful extraction of high-density microplastics such as polyvinyl chloride (PVC) (density: 1.16–1.58 g/cm^3) and polyethylene terephthalate (PET) (density: 1.37 1.45 g/cm^3) (Prata et al., 2019a).

Pre-treatment methodology is summarized in Fig. 2. Briefly, the influent of DWTP was filtrated through stainless-steel sieves (Cisa Sieving Technologies, 200 mm diameter x 50 mm height) with mesh sizes of 20, 50, 100, 500, and 1000 µm. Each fraction was filtered through vacuum filter equipment on a 10 µm PTFE filter, treated with Fenton's reagent, and sonicated for 10 min at 40 °C. The suspension was left for 24 h and was filtered again through a 10 µm PTFE filter. Density separation with zinc chloride solution (1.8 g/mL) was performed next. The sample was sonicated at 40 °C for 10 min and left for 4–12 h. Furthermore, it was centrifugated at 3000 rpm for 1 min and the supernatant was filtered through a 10 µm PTFE filter. The process was repeated three times on the same filter. The quantity of organic and inorganic matter, in the sieves with particle size ≥50 µm was not significantly high to interfere with MPs identification and for this reason, visual inspection was performed immediately and the oxidation process for organic matter removal was not carried out. In contrast, for particles between 20 and 50 µm, the PTFE filters were treated with hydrogen peroxide 33–35% for 24 h.

Samples of dry sludge and water from sludge centrifugation units were filtrated through stainless-steel sieves with mesh sizes of 20, 50, 100, 500, and 1000 µm in the laboratory and dried at 40–45 °C for 3–4 days, and the weight of dry material was recorded. The methodology for processing was elaborated with a combination of steps and different sequences based in Mintening et al.,(2017), Prata et al., (2019b), Ben-David et al., (2021), Ziajahromi et al., (2021), Elkhatib and Vinka Craver (2020) methodologies. In this study Fenton's solution was applied for 24 h due to high organic matter content.

For clarifier lines 1 and 2, sand and carbon filters as well as treated water units of DWTP, due to low organic and inorganic particle content on the PTFE filters, were directly inspected taking into account MPs size

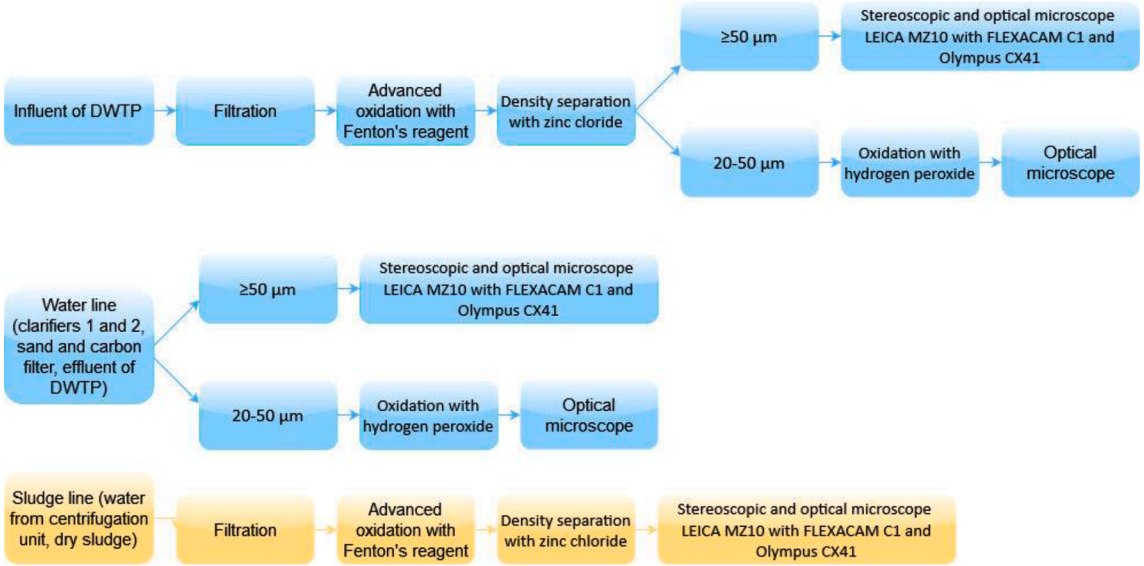

Fig. 2. Pre-treatment protocols for microplastic identification and visual inspection.

equal to or larger than 20 μm.

Visual identification was performed for all filters of treatment units and after purification of the river and sludge samples with a stereoscopic microscope LEICA MZ10 with 1 μm resolution and 80 x magnification coupled to FLEXACAM C1 camera and docked screen of 32 cm, light projected from above and below to get good image of surface structure and appearance, in order to sort the particles according to their most common shapes: fibers, films and fragments (Bayo et al., 2020; Hidayaturrahman and Lee, 2019; Lares et al., 2018) as well as to quantify them. Particle shapes between 20 and 50 μm were then confirmed with an Olympus CX41 optical microscope at 40 and 50X magnification and 400 and 500 X total magnification. All particles identified were extracted from the filters and were taken to analysis for composition

In order to determine the size of the MPs, ImageJ software was used, previously calibrated with *Olympus Objective Micrometer* reference (10 μm). Fiber length was registered and for fragments the mean between maximum and minimum diameter was registered.

2.4. Qualitative analysis

After visual identification, MPs composition was analyzed by μFTIR and ATR-FTIR spectroscopic techniques. Only for river, water from sludge centrifugation and sludge samples, of the total number of particles between 20 and 500 μm, sub-samples were extracted on calcium fluoride slides for maps elaboration due high quantity of possible plastic particles. For subsample, particles were chosen according to morphology, color, and appearance for every filter section, until completing the entire filter area to ensure representativeness. For other samples (clarifiers, sand, carbon filter, and effluent), all particles detected were analyzed for their composition.

Particles larger than 500 μm were analyzed by Thermo Scientific GladiATR Highest Performance NICOLET Diamond ATR-FTIR spectrometer with OMNIC™ Paradigm Software. The measurements were performed in reflection mode in the range of 400–4000 cm^{-1} with 16 scans at a spectral resolution of 4 cm^{-1}. The background was done before analysis and every 6 samples. Infra-red maps over calcium fluoride slides were performed for particles from 20 to 500 μm by μFTIR analysis or ultrafast mapping microscope Thermo Scientific Nicolet™ iN™10 with MCT detector with pixel aperture 25x25 μm in transmission mode, in the range of 715–4000 cm^{-1} with 4 scans at a spectral resolution of 4 cm^{-1}. The spectra were analyzed with the OMNIC Specta MCS Software and the identification was obtained by comparing unknown spectra with databases such as HR Nicolet Sampler Library, Hummel Polymer Sample Library, Polymer Laminate Films, Wizard Library as well as an own library that was generated with more than 80 spectra.

For all particles isolated from the treatment unit's samples such as clarifiers 1 and 2, sand filter, activated carbon filter, and treated water, all particles ≥20 μm were placed over calcium fluoride slides for μFTIR analysis using Leica stereoscopy microscope (resolution of 1 μm).

To avoid the loss of MPs, for sludge, water from sludge centrifugation and river samples, in the PTFE filters containing particle sizes from 20 to 50 μm, all particles were retired from the filter for composition analysis, then for missing particles recruitment, the filter was washed vigorously with hydrogen peroxide 33–35% for 24 h and filtered on siliceous filters (1 cm^2, pore size 5–6 μm wide, square shape, 12 μm pore distance, membrane thickness 500 μm). The entire filter was checked first for some single particle presence with microscopy μFTIR and then μFTIR maps were elaborated. For μ-FTIR maps, an area of 3 mm^2 was established randomly over siliceous filters considering interference absence and accurate IR spectra generation, in that area, a spectrum was made every 25 μm with a total of more than 15,000 spectra for analysis with Omnic Picta Software and libraries selected as HR Nicolet Sampler Library, Hummel Polymer Sample Library, Polymer Laminate Films, Wizard Library. The μ-FTIR maps were obtained by reflection, four scan accumulations, the spectral resolution of 4 $cm^{-1,}$ and spectral range of 4000–715 cm^{-1}.

Similarly, all PTFE filters from all treatment units of DWTP were also treated with hydrogen peroxide 33–35% for 24 h and filtered over siliceous filters. All siliceous filters were analyzed as above mentioned.

Match spectra greater than or equal to 70% similarity to the reference spectra were accepted. Unidentified spectra were compared to two own libraries elaborated with more common plastics in the market and weathered fragments, films, and fibers from environmental samples applying the same criteria for reference spectra acceptance. Furthermore, particles were analyzed according to their characteristic absorption band of each polymer chemical grouping bonds according to Campbell et al., (2000) and Coates (2000), for frequencies ranging mostly from 3650 to 500 cm^{-1}, 1480–1430 cm^{-1} for C—C aromatic ring stretching, 1790–1700 cm^{-1} for double binding C—O stretching, 2980–2780 cm^{-1} for C—H stretching of aliphatic and 3150–3030 cm^{-1} of aromatics. For aliphatic organohalogen, natural and synthetic cellulose identification, frequencies from 1150 to 550 cm^{-1}, 1200–1000 cm^{-1}, 1500–1200 cm^{-1} (C—H and C—O—H bending modes), 2900–2820 cm^{-1} (C—H stretching modes) and from 3600 to 3000 cm^{-1} (O—H stretching modes) were also evaluated. The particles identified as plastic were added to the final quantification. If the particle still could not be identified, it was considered to be a definite unidentified category.

2.5. Quality control

To avoid cross-contamination, ultrapure water was filtered through a 0.45 μm pore size filter (*GF/F Whatman*, 47 mm diameter, nitrocellulose). All equipment and materials were washed with filtered ultrapure water and 70% ethanol before sampling. All reagents were prepared with filtered ultrapure water. During the analysis of the samples in the laboratory, only laboratory coats made of cotton were worn. The extraction hoods were used during all sample pre-treatment processes. Moreover, procedural blank samples were collected in the field as well as in the laboratory and the corresponding corrections were applied to the results.

Controls were applied during river sampling activities and consisted of two open Petri dishes, 90 cm in diameter, to collect possible airborne fibers. The results were 8 fibers (3 black, 2 red, and 3 blue) and 6 fibers (3 black, 2 blue, and 1 red) in control 1 and 2, respectively. In consequence, the maximum fibers value of 8 samples were removed of the final quantification result of the various PTFE filters analyzed from every sieve size used in river water filtration. This selection was in a randomly way, choosing several filters and removing the fibers according only by color and length. Inside the laboratory of DWTP, for catchment samples, were located two controls near of sampling pipelines that representing the outlet of each treatment unit, two 60 mm petri dish was placed open near the sampling area while activity was developed. The results were 0 particles in both controls.

For accurate results and due to the sample processing site in laboratory is not entirely free from fibers a deposition rate of airbone fibers was calculated prior to sample processing. This rate was calculated for the laboratory workspace where the samples were processed. The procedure was four open petri dishes (60 cm in diameter) with PTFE filters were placed into similar distance in laboratory and covering the entire area for a month, after that the fibers deposited were quantified and referred by filter area and time with a result of 0.04 fibers/cm^2/day. Considering a PFTE filter area of 17.4 cm^2, 1 fiber was randomly subtracted from each analyzed filter needed organic matter or turbidity removals. Finally, in the procedural blank, MPs (fibers, films, or fragments) were not found.

For the detection of particles lost through the different steps of the sample treatment, a recovery test ($n = 3$) was conducted per sample batch, spiking a mix of colored polyethylene spheres with diameters from 53 to 500 μm (Cospheric Inc., California, USA) in water and sludge samples. The recovery rates in sludge were 75 and 91% for 53–63 μm, 74 and 95% for 125–150 μm, 70 and 95% for 250–300 μm, and 100 and

100% for 425–500 μm for sludge and water samples. No degradation signs (i.e. mechanical damages, fragmentations, or chemical attacks) were observed on spheres. The methods applied in the present work fulfill almost all minimum requirements established by Schymanski et al. (2021).

3. Results and discussion

3.1. Abundance and removal of microplastic particles in the water line of DWTP

Microplastic particles were detected in all water samples, from influent to effluent. Table 1 presents the MPs concentrations, removal efficiency, and sizes for each treatment unit in the water line. Due to the morphological characteristics of the fibers and their large length/width ratio, they were not categorized in size fractions, yet their size can be found in Supplementary data.

The concentration of MPs in the DWTP influent (raw water) was 4.23 ± 1.26 MPs/L. The most abundant MPs concentration was between 500 and 100 μm (43%) followed by a fraction of 100–50 μm (30%) considering films and fragments. These results are lower than the results published by Pivokonský et al. (2020) whose concentration, for raw water, was 1296 MPs/L and 23 MPs/L, for advanced and conventional treatment, in the DWTP located in the Czech Republic, respectively. The different results are mainly caused by different size ranges, as the previous study investigate MPs up to 1 μm. Another study by Mintenig et al. (2019) determined concentrations of MPs, larger than 20 μm, in raw water, that ranged from 0 to 0.007 MPs/L, although raw water refers to groundwater extraction at least 30 m deep, in Germany. Other studies conducted in European rivers found MPs mean levels of 12, 42, and 5.53 MPs/m^3, in Rhone, Tet, and Elbe river, respectively (Constant et al., 2020). However, influent water cannot be representative of river water, and water from the Mediterranean river, here studied, are not comparable to the rivers of northern or central Europe. Only a recent study (Dalmau-Soler et al., 2021) was found regarding the same river that feeds the DWTP here studied, with similar MPs levels. Along the river basin, concentration was ranging from non-detected to 3.6 MPs/L while the mean concentration in the influent of DWTP was 0.96 ± 0.46 MPs/L. Still, data on the presents of MPs in water in Mediterranean rivers, is very scarce (Guerranti et al., 2020). However, the differences between methodologies, lack of standardization, and size ranges of analyzed MPs make difficult these comparisons.

The overall MPs removal rate was 98.3%, from 4.23 ± 1.26 MPs/L, in raw water, to 0.075 ± 0.019 MPs/L in the effluent. Considering each treatment unit, clarifier eliminated the most of MPs, between 63 and 67%, depending on the water line studied. The most abundant size fraction for both clarifiers was between 500 and 100 μm considering films and fragments. Subsequently, the sand filter increases the removal efficiency up to 99% where the concentration of 0.038 ± 0.025 MPs/L was found in the effluent of this treatment unit. Previous research by Dalmau-Soler et al. (2021) and Li et al. (2020) also reported that the first treatment stage (coagulation/sedimentation in clarifiers and sand filtration) had the highest impact on MPs removal, investigating full-scale DWTPs, as well as some lab-scale studies by Ma et al. (2019). Furthermore, a recent study by Cherniak et al., 2022 found that coagulation, flocculation, and sedimentation accounted for the highest removal (70%), however negligible additional removal was observed in the filtration unit (< 1%).

The next treatment unit, the activated carbon filter, does not increase significantly the removal efficiency with a level of 0.032 ± 0.025 MPs/L in their effluent. By contrast, after the microfiltration, electrodialysis, and remineralization stage, an increase of MPs content, from 0.032 to 0.075 ± 0.019 MPs/L, was observed. Wang et al. (2020) observed similar behavior in the effluent of the ozonation unit, leading to an increase in small and fibrous MPs. The reason may be that the MPs in this operating unit broke down due to the shear force of the water flow (Horton et al., 2017). Johnson et al. (2020) noted that there is a possibility of MPs generated within DWTP itself, while Dalmau-Soler et al. (2021) performed migration tests to check this theory, and the results suggested that MPs can be released from DWTP when the materials are old; however, this probably does not occur at normal DWTP working conditions. Our speculation was that remineralization of the treated water with calcium hydroxide and post-chlorination with sodium hypochlorite probably added particles to the effluent water. These MPs pollution could come from solid reagents ($Ca(OH)_2$) packaged with plastic, chlorination, or pollution coming from the resuspension of storage tanks, however more studies are needed to elucidate this. Nevertheless, the removal efficiency of DWTP, in this study (98.3%), is higher than the ones observed in other European plants that ranged from 70 to 90% (Pivokonsky et al., 2018; Pivokonský et al., 2020). Other work performed in a pilot station established a removal rate of MPs fibers and fragments in 81 and 89%, respectively, in absence of coagulant and 96 and 97%, respectively, in presence of coagulant (Negrete Velasco et al., 2022). When comparing different water treatment plants in the same river basin, the removal rate reported was similar with an overall value of 93% (Dalmau-Soler et al., 2021). Certainly, direct comparison between the studies should be taken with caution due to differences in DWTP technology, MP sampling, sample pre-treatment, analytical identification methods to differences related to spatial variations and sampling location (Novotna et al., 2019).

Regarding the DWTP effluent, the concentration was 0.075 ± 0.019 MPs/L (fraction between 100 and 50 μm most abundant considering films and fragments), which is higher by one order of magnitude than the average value determined in a German DWTP that catch underground water (Mintenig et al., 2019) of 0.007 MPs/ L. Potable water from eight DWTPs within the UK had similar results, typically less than 0.002 MPs/L, accounted for MPs ≥ 25 μm (Johnson et al., 2020) and also similar to effluent water of Spanish DWTP located in the same river basin (Dalmau-Soler et al., 2021), with MPs levels ranging from 0.03 to 0.11 MP/L. On the contrary, these values were much lower than the values reported by Wang et al. (2020) of 930 ± 71 MPs/ L in the effluent of one of the largest advanced drinking water plant located in China. The typical capacity of this plant is 120 million m^3/day compared to the treatment plant in our study, with a treatment capacity of only 276,480 m^3/day. A recent study investigated tap water samples from different developed countries including France, Germany, Japan, and the United States, showing much higher results than the ones obtained in the present study, with the MPs concentration ranging from 1.9 to 225 MPs/L,

Table 1
Levels (MPs/L), size distribution and removal efficiency of MPs in the DWTP.

Unit	*n*	Concentration [MPs/L]	Removal efficiency [%]	Size (μm)*				
				2000–1000	1000–500	500–100	100–50	50–20
DWTP influent	3	4.230 ± 1.263	-	6%	13%	43%	30%	8%
Clarifier line 1	7	1.577 ± 0.696	62.7	<1%	30%	50%	20%	<1%
Clarifier line 2	7	1.388 ± 0.543	67.2	<1%	<1%	50%	25%	25%
Sand filter	3	0.038 ± 0.025	99.1	<1%	<1%	67%	33%	<1%
Carbon filter	3	0.032 ± 0.013	99.2					
DWTP Effluent	7	0.075 ± 0.019	98.3	<1%	<1%	39%	46%	15%

*Size was analyzed for films and fragments. Concentration expressed as mean standard ±deviation

and the overall mean concentration of 39 ± 44 MPs/L (size range: 19.2 μm to 4200 μm) (Mukotaka et al., 2021). Kosuth et al. (2018) also investigated MPs in the tap water from 14 different countries in Europe, America, Asia, and Africa with the concentration ranging from 0 to 61 MPs/L and the overall mean of 5.45 MPs/L (size range: 100 to 5000 μm). The highest concentration of 61 MPs/L was found in the USA, while the four lowest means were from European Union countries: non-detected; 0.91; 1.82; 1.83 MPs/L for Italy, Germany, France, and Ireland, respectively. However, it should take into account the low volumes (0.5 L) of water analyzed by this study (Kosuth et al., 2018). In some cases, MPs concentration could vary from the effluent of DWTP to tap water, since the household pipes made from durable plastic (PVC, PE, and PA) could contribute to the increase of MPs in the tap water over time (Mintenig et al., 2019; Tong et al., 2020), however, Shen et al. (2021) reported no difference between these two samples.

The morphology of MPs has also been investigated and visually categorized into three main shape types: fragments, films, and fibers (Bayo et al., 2020; Hidayaturrahman and Lee, 2019; Lares et al., 2018). Fig. 3 shows that fiber morphology was the most prevalent in DWTP influent, similarly reported by other authors (Cherniak et al., 2022; Dalmau-Soler et al., 2021; Sarkar et al., 2021; Wang et al., 2020).

The proportion of fibers decreased while treatment advanced from 77% (MPs/L) in raw water to 27% (MPs/L) in the effluent. Several explanations are present regarding the better removal of fibers, especially in the first treatment steps. Fibers are more likely to form flocs and settle (Katrivesis et al., 2019) since they are usually heavier polymers such as polyamide (PA) or nylon (density 1.15 g/cm3), polyester (PES) (density 1.38 g/cm^3) and cellulose acetate (CA) (density 1.30 g/cm^3) and their chemical structure can facilitate attachment to coagulant flocs such as carbonyl (C═O) chemical groups of PES or PET (Xue et al., 2022), also PA fibers can be ionized depending on pH media and interact with coagulants and flocculants. Furthermore, the surface of MPs has an impact on its removal, so the rougher the surface, the better the removal (Shahi et al., 2020). It is most likely, that these synthetic fibers, in the water environments, are released from washing machines (Napper and Thompson, 2016). Fragments and films are microplastics that remain in the effluent water and are more difficult to remove during the treatment process with an abundance of 47 and 26%, respectively. Fig. 4 shows different morphology types detected in the influent and effluent of DWTP.

3.2. Abundance of microplastic particles in the sludge line of DWTP

Microplastic removed from the water, during the treatment process, tends to accumulate in the sludge (Conesa and Ortuño, 2022). Dry sludge and water from sludge centrifugation unit MPs concentration are depicted in Table 2.

Water from the sludge centrifugation unit had a MPs concentration of 194 MPs/L dominated by particles of sizes ranging from 2000 to 1000 μm (Fig. 5A). Fibers were more than 75% of MPs detected in water from the sludge centrifugation unit. The concentration in the dry sludge was 14,360 MPs/kg$_{w.w.}$ (or 35,680 MPs/kg$_{dw}$). The highest concentration of particles was between 500 and 100 μm, followed by a fraction of 100–50 μm (Fig. 5B). Fibers were, again, the most abundant morphology in general, however, it can be observed that their concentration decreases from larger size ranges (2000–1000 μm) to the smallest (50–20 μm), where film and fragment type increases significantly. High fiber content was in line with high removal efficiency from treated water. Another study also collected sludge produced during the water treatment process, at four DWTPs in the UK. The results varied extremely from non-detected to 86,000 MPs/g $_{dw}$. The authors commented that more samples are necessary to understand such variations (Johnson et al., 2020).

Taking into account the results obtained in our study, we suggest that both dry sludge and water from sludge centrifugation should be properly managed as waste, avoiding agricultural uses or its return to the river, in order not to transmit contamination to terrestrial ecosystems. The implementation of new technologies that enable the removal of MPs from sludge as well as from environmental samples is definitely needed in order to move forward towards a circular economy for plastic waste and to protect the environment (Sol et al., 2020).

3.3. Composition of microplastic particles

Each sample, after visual inspection and particle quantification, was photographed with Leica FLEXACAM C1, IC90 E, ICC50 W/E camera and subject to final confirmation with FTIR spectroscopic technique. For this purpose, MPs maps were elaborated. In Fig. 6 can be seen μFTIR map for the influent of DWTP.

A total of 345 particles were confirmed and 22 different polymers were identified; however, the most common were: synthetic-cellulose, PES, polypropylene (PP), PA, and polyethylene (PE) (Fig. 7). In addition, PET and polystyrene (PS) were also found. Pivokonský et al. (2020) reported similar results for the most commonly observed MPs in almost every sample: CA, PET, PVC, PE, and PP. Synthetic cellulose (rayon,

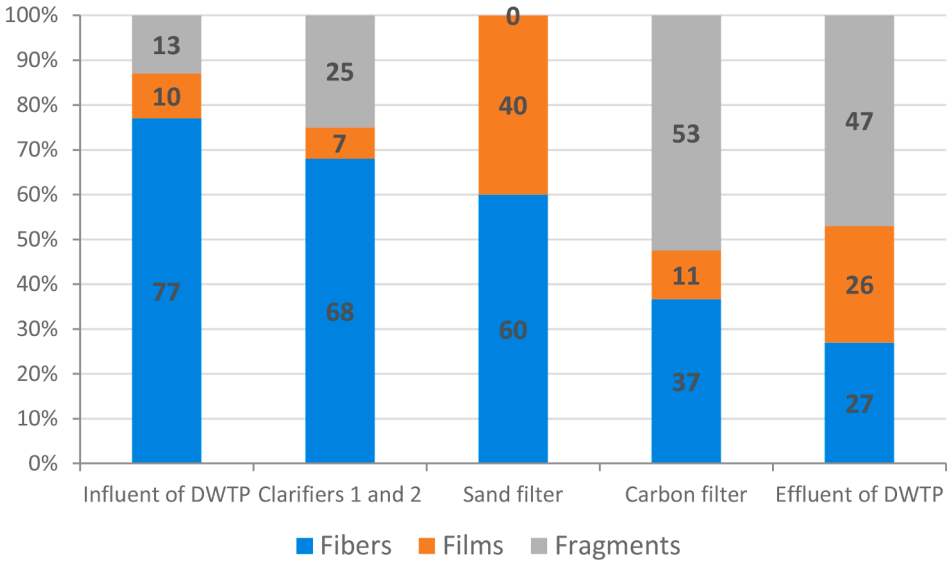

Fig. 3. Microplastics morphology for different treatment units of drinking water treatment plant (DWTP).

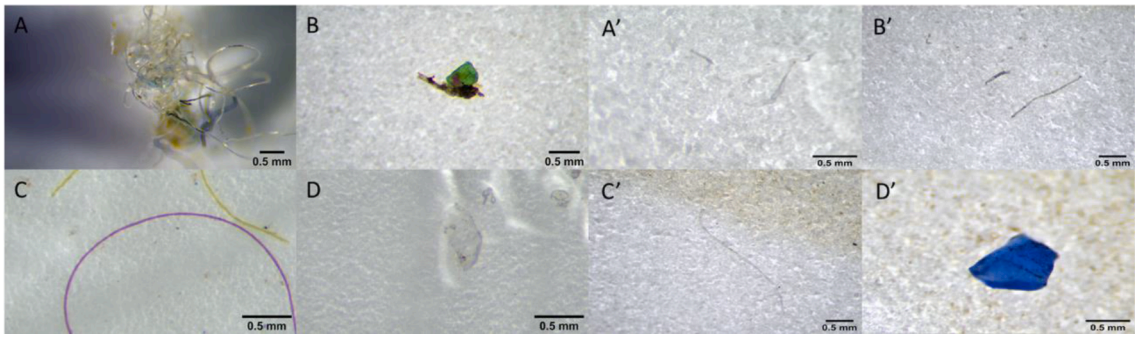

Fig. 4. MPs detected in influent: (A) group of fibers (B) fragment (C) individual fibers (D) films and effluent of DWTP: A'. film and fiber B'. fibers C'. fiber D'. fragment with 50–80 x magnification.

Table 2
Concentration of MPs and size distribution in the sludge line of the DWTP.

Unit	Concentration [MPs/L][MPs/kg$_{w.w.}$]	Size (μm)*				
		2000–1000	1000–500	500–100	100–50	50–20
Water from centrifugation	194	52%	<1%	7%	13%	28%
Dry Sludge	14,360	24%	10%	32%	26%	8%

*Size was analyzed for films, fragments and fibers.

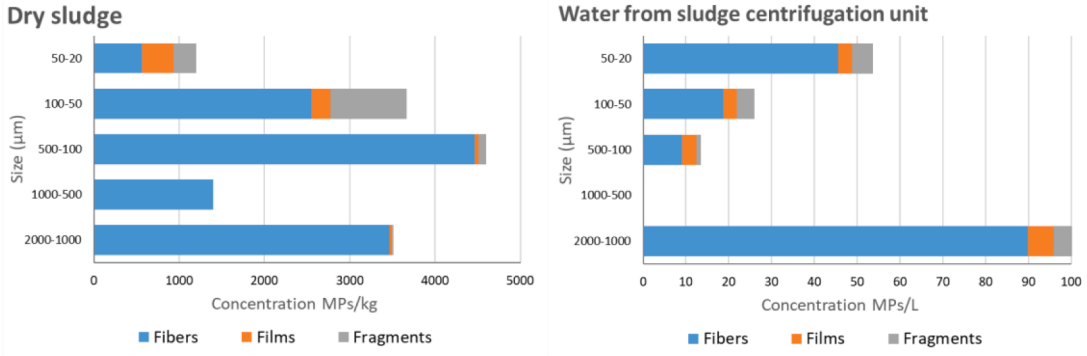

Fig. 5. Microplastics concentration and morphology for each size in dry sludge and in water from sludge centrifugation unit.

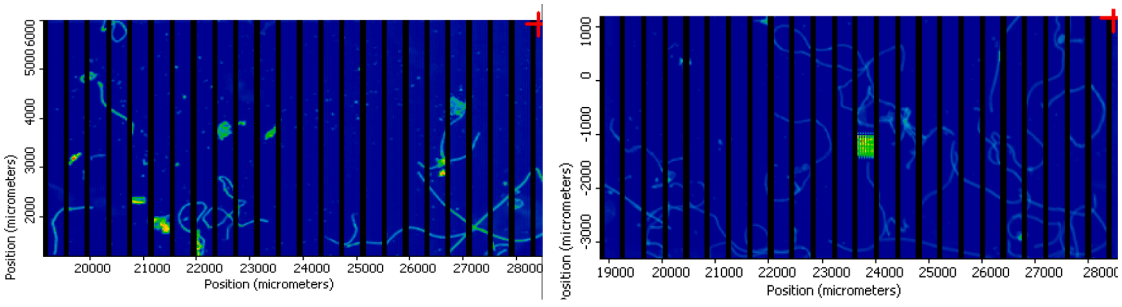

Fig. 6. microFTIR maps for influent of DWTP.

cellulose acetate) was found in all samples with a high abundance from 24 to 53%. Although some authors, in previous studies, reported it as a non-plastic particle (Wang et al., 2020; Yu et al., 2016) in the present study it was considered a synthetic polymer and was included in the quantification of the microplastics. One of the reasons was that after pre-treatment of the samples, cellulose did not degrade, which should happen in the case of natural material, indicating synthetic origin. All of these polymers are commonly used materials. Cellulose, PES, and PA are synthetic fibers used in the textile industry; PE is used for reusable bags, trays, containers, toys, and household utensils, while PP is used for food packaging, pipes, car parts, etc. (PlasticEurope, 2021).

Other polymers detected in this study were: polyurethane (PU), PET, polyacrylonitrile (PAN), Tygon B 44–4 polymer (a new generation polymer with high content of PVC and silicon, but without phthalates), with abundance >5% (which are included in Fig. 7) and polycarbonate (PC), PS, PVC, polybutadiene (PBD), polyisobutene (PIB), polymethyl methacrylate (PMMA), polyvinyl acetate (PVA), polyvinyl stearate, PP-PE copolymer, epoxy resin, alkyl-resin, ethylene vinyl alcohol/ethylene vinyl acetate (EVOH/EVA), fluoroelastomer (synthetic rubber) with abundance <5% (the proportions of materials with low abundance in

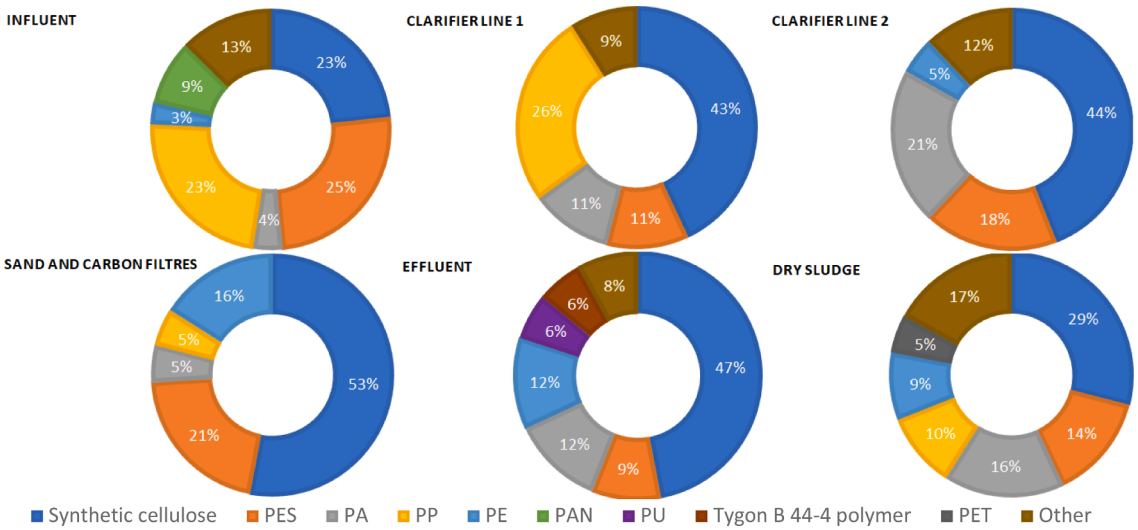

Fig. 7. Composition of microplastic (films, fragments and fibers) from different units of the drinking water treatment plant. PES (Polyester); PA: Polyamide; PP: polypropylene; PE: polyethylene; PAN: polyacrylonitrile; PU: polyurethane; PET: Polyethylene terephthalate.

DWTP units <5% are denoted as "other").

Considering MPs composition and morphology, fibers in the water line were made of synthetic cellulose, PES, PA, PAN, and to a lesser extent of PMMA, while fragments and films are mainly made of PP, PE, PA, and to a lesser extent of PU, polyvinyl stearate, PP-PE, and PET. For dry sludge, besides the main fiber composition (synthetic cellulose, PES, and PA), fibers made of PAN, PE-PP copolymer, and PP were also detected to a lesser extent. Fragments and films in sludge were made of PE, PP, PA, and to a lesser extent PET, PVC and PU.

Some polymers that are detected in the effluent of DWTP were not found in the influent, opening the possibility of MPs generated within DWTP itself. However, to confirm this statement, more proofs are needed. Due to the COVID-19 pandemic and the global use of disposable masks that are consisting of polymers such as PES, PP, and PS, in the following years is expected a significant increase in its concentration in freshwater sources (Xu and Ren, 2021).

According to the present results, assuming a water consumption per capita of around 1.0 L/day (Martínez et al., 2017), all tap water coming from this DWTP, and no extra incorporation of MP in distribution pipelines, the annual intake of MP through drinking tap water was set at 27 MP/person/year. This MPs intake through drinking water is negligible compared with another study that set the annual ingestion of MPs, via food and beverages, between 39,000 and 52,000 items/year (Cox et al., 2019). Most probably, seafood is one of the top three contributors to MPs human consumption, especially in the countries like Japan, with a much greater dietary proportion of seafood, with the highest estimation of 104.2 g/day, leading to daily consumption of 154 MPs (Cox et al., 2019; Micha et al., 2015). Another study (Catarino et al., 2018) indicated that MPs ingestion by a human via wild mussels is minimal compared to exposure via household fibers fallout during a meal. They predicted a concentration of 123 MPs/year/capita in the UK for MPs ingestion by humans via consumption of mussels and it can go to 4620 MPs/year/capita in countries with higher shellfish consumption such as France, Belgium, and Spain. By comparison, fiber exposure during a meal via dust fallout in a household was estimated to be 13,731–68,415 MPs/year/capita.

4. Conclusions

Microplastic particles are detected in all samples within DWTP but their concentration varies significantly. In general, the operation of a drinking water treatment plant with respect to the reduction of microplastic particles is effective and with elevated removal efficiency. Clarifiers together with the sand filtration unit had the greatest impact on MPs reduction, >99%. However, an increase in microplastic particles, in the DWTP effluent, was observed and the final removal efficiency was 98.3% which is still higher than the values reported for other European drinking water treatment plants. It should be noted that direct comparison between the papers is challenging, since to date, there is no standardized analytical method for MPs identification.

As observed in the case of wastewater treatment plants, microplastic particles that are removed during treatment processes tend to accumulate in the sludge, especially synthetic fibers that are coming from washing machines. Both sludge and the centrifuged water must be properly managed as waste, avoiding agricultural uses or its return to the river, not to transmit contamination to terrestrial and aquatic ecosystems. New technologies should be implemented that enable the MPs removal from the sludge.

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

Data will be made available on request.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.watres.2022.119185.

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