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Evaluation of microplastics removal efficiency at a wastewater treatment plant discharging to the Sea of Marmara☆

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ABSTRACT

Levels, composition and fate of microplastics (MPs) were investigated along different compartments of a secondary wastewater treatment plant (WWTP) with nutrient removal on the northern Sea of Marmara coast (Istanbul, Turkey). When all samples were combined, fibers were found to be the most dominant particles, followed by hard fragments. 500–1000 μm and 1000–2000 μm were the most common size ranges for wastewater and sludge, respectively. Rate of removal differed for sizes and shapes of the particles combined. Hard fragments of <500 μm and fibers of size ranges 250–500 μm and 1000–2000 μm were more successfully removed within the WWTP. Size averages increased throughout the WWTP units. 84.6–93.0% removal was achieved for grab and 3-hr composite samples. Despite the high removal rates of the WWTP, 2,934 × 10^6 microplastic particles/d were released in the effluent to the Sea of Marmara. Our results show that the Ambarlı WWTP considerably contributes to microplastics contamination in the Sea of Marmara since the plant has a high operating capacity.

1. Introduction

Plastics are now a ubiquitous component of waste stream, due to leakage of many commercial products containing plastics. Plastics have been detected in rivers, lakes and oceans (Cole et al., 2011; Güven et al., 2017; Kideys and Aydın, 2020) as well as in soil and air (Dris et al., 2016; Steinmetz et al., 2016; Horton et al., 2017). Particles <5 mm are classified as microplastics, a scale commonly used in the literature (Barnes et al., 2009; Duis and Coors, 2016). Microplastics can further be categorized as primary and secondary microplastics (Akdogan and Guven, 2019).

Primary microplastics mostly originate from personal care products, drugs and pellets used for production of plastic consumer products (Fendall and Sewell, 2009; Patel et al., 2009; Sundt et al., 2015; Browne et al., 2011; Napper et al., 2015). The microplastics present in personal care products may enter aquatic environments through effluent discharges of wastewater treatment plants (WWTPs) (Duis and Coors, 2016; Anderson et al., 2017) and run-off or mismanagement during production or storage of pellets used in various products including personal care products (Sundt et al., 2015). Secondary microplastics are the fragments arising from the breakdown of larger plastics at sea or land (Cole et al., 2011), due to physical, chemical and biological processes or exposure to UV radiation (Barnes et al., 2009; ter Halle et al., 2016; Okoffo et al., 2019). Fibers are the commonest type of secondary microplastics and they are most likely to be generated during the washing of synthetic clothing (De Falco et al., 2018; Hernandez et al., 2017; Napper et al., 2015).

WWTP effluent discharges are one of the major point sources of microplastic pollution into the environment, especially for primary microplastics and fibers since they share the same pathways into the environment (Horton et al., 2017). Sludge generating from WWTPs also contains microplastics that are removed from wastewater during purification (Li et al., 2018; Okoffo et al., 2020a). Sludge generated from WWTPs, under certain conditions, can end up in soil for agricultural and landscaping purposes as well as landfills (Ng et al., 2018; Corradini et al., 2019; Okoffo et al., 2020b).

Microplastics discharged from WWTPs are transported to many areas of the environment. Through rivers and water channels, they end up in the sea or in the ocean (Jambeck et al., 2015) or deposited in sediments (Nel et al., 2018; Besseling et al., 2017). Through application on land, microplastics may enter groundwater, via rainwater they reach rivers and seas, and carried in the wind, plastic particles are further

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transported overland (Zylstra, 2013; Dris et al., 2016; Akdogan and Guven, 2019).

WWTPs receive several types of influent streams depending on sewer infrastructure. A plant can receive solely domestic wastewater if a separate discharge system is applied, if a combined discharge system is utilized, the plant receives storm water runoff and partial industrial wastewater in some cases (Ngo et al., 2019). Although the removal rates of microplastics are generally high in WWTPs, they still discharge considerable amounts of microplastics in their effluent streams with high flow rates (Michielssen et al., 2016; Murphy et al., 2016; Edo et al., 2020). Sludge produced in WWTPs also contains high quantities of microplastics (Leslie et al., 2017; Edo et al., 2020; Liu et al., 2019).

Gündoğdu et al. (2018) investigated two WWTPs in Adana, Turkey and Akarsu et al. (2020) investigated three WWTPs in Mersin, Turkey for microplastics content in influent and effluent streams. Bilgin et al. (2020) examined the presence of microplastics in influent and effluent streams as well as aerated grit chamber effluent and sludge at a WWTP in Sakarya, Turkey. Daily capacities of the investigated facilities ranged between 12,000 and 190,000 m^3/day. From the available literature, we encountered no study investigating the WWTPs located along the Sea of Marmara. Istanbul (Turkey) is the largest city located in the northern region of the Sea of Marmara and western area of the Black Sea, which being the most populated city in Europe, produces vast quantities of wastewater. There are 87 WWTPs in Istanbul discharging mainly to the Sea of Marmara including the Bosphorus Strait and also to the Black Sea with an overall capacity of 5,881,660 m^3/d (Figure S1). The aim of this study was to evaluate efficiency levels of various compartments at a high capacity advanced WWTP in Istanbul to better understand the removal dynamics of microplastics.

2. Materials and methods

2.1. Description of investigated facility and sampling points

Samples were collected from the Ambarlı WWTP, Istanbul, Turkey serving around 2 million people. This secondary WWTP with advanced nutrient removal possesses a total capacity of 400,000 m^3/d, an average effluent discharge of 360,083 m^3/d and dried sludge production of 100.7 tons/d. Effluent from the plant reaches the Marmara Sea via a water channel. Wastewater samples were collected from the entrance of the WWTP (1), effluent of the grit/grease removal chamber (2), effluent of the aeration basin (3) and final effluent of the WWTP (4). Sludge sample was collected from dryers after digestion of the sludge (5) (Fig. 1). Throughout this study, obtained samples are abbreviated as: Influent sample (IN), physical treatment effluent (PHG for grab sample and PHC for 3-hr composite sample), biological treatment effluent (BIO), final effluent sample (EFG for grab sample and EFC for 3-hr composite sample) and dried sludge sample (SLD), respectively. The overall wastewater samples are abbreviated as WW and samples collected from all WWTP units are labelled TOT.

2.2. Sampling and extraction

Wastewater samples were obtained by grab sampling conducted with steel buckets samples were collected into glass bottles. Sludge sample was collected with a glass jar directly from the dried sludge pipeline. Sampling was conducted on December 26, 2019. Wastewater sample volumes were 5 L for raw wastewater, 10 L for physical treatment effluent, and 1 L for biological treatment effluent, due to presence of different amounts of visible microplastics and organic compounds in the samples. Specifically, BIO and IN samples contained significant amount of organics, for which lower volumes were used not to damage plastics during digestion and extraction processes within the contact times. Two identical streams were present in the WWTP. 30 L of final effluent was sampled as a mixture of 15 L from each stream. Physical treatment effluent and final effluent were additionally sampled as 3-hr composite samples whereby samples were collected every 15 min over a 3-h period, in order to understand fluctuating streams. Dried sludge was sampled at a volume of 0.25 L. Samples were kept at 4 °C for a maximum of 7 days before pre-treatment and extraction.

Depending on the organic content of the samples, extractions were conducted by a modified version of the recommended wet peroxide oxidation method from Lares et al. (2019) and method proposed by Nuelle et al. (2014). If suspended solids contents were high and the color of the suspended solids were yellow-brown or brown, the sample was considered to have a high organic content. Samples were sieved through 25 μm-2mm sieves and 50 μm meshes were used for concentrating the particles, as opposed to 20 μm–250μm sieves and 0.8 μm meshes used in Lares et al. (2019). For SLD and BIO samples with high organic content, sequential digestions were carried out if the organic content was still high and no reactions observed due to maximum volumes in the reacting flask. The mixture was left to settle after which the aqueous phase was filtered off and subsequently the digestion for the settled fraction was performed with the same parameters as the preliminary digestion stage. Overall, reaction times were no more than 7 days for all the methods applied to all sample types.

2.3. Microplastics identification and characterization

In general, the standard EC guidelines on the processing of microplastic samples were followed (European Commission, 2013). Microplastics were visually investigated under a stereo microscope (Olympus SZX 16, 30× magnification). Particles identified as MPs were separated using micro tweezers into a clean Petri dish containing a clean cellulose filter paper, previously checked for airborne contamination. The color and shape of each microplastic particle was recorded and photographed under the microscope.

The shapes included mainly fibers, hard and soft fragments, and others (i.e. pellets, glitter and rubber). Using the forceps, slight pressure was applied to MP particles and depending on their reaction to applied pressure fragments were classified as hard or soft fragments (Güven

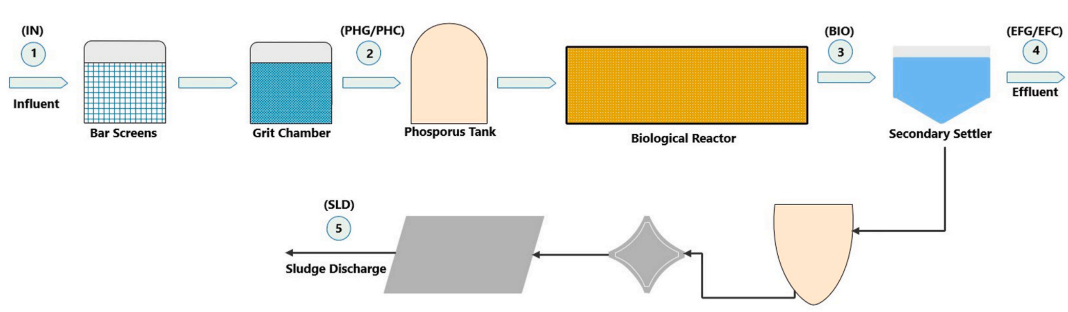

Fig. 1. Flow diagram of WWTP and sampling points (Modified from Akarsu et al., 2020).

et al., 2017; Akarsu et al., 2020). Particles displaying hexagonal, notched morphology with straight edges and measuring circa 250 μm were classified as glitter (Yurtsever, 2019). Pellets (or microbeads) generally have spherical shapes and equal dimensions. ImageJ software was employed for measuring maximum length of particles.

The separated particles were referred to as microlitter (ML). Microlitter is a term used to describe particles found in water samples, which are smaller than 5 mm and of unnatural origin. The term is also used to describe the particles suspected as MPs where no characterization data is presented. Polymer characterizations were conducted with a Raman Microscope (Renishaw Invia) using 532 nm and 785 nm lasers. Spectra were recorded in the 200-3400 cm^{-1} wavenumber range, using 20× objective and laser powers between 0.5 mW and 10 mW. Integration time was 100–200 ms and 10–20 scans were accumulated. BioRad KnowItAll ID Expert Software with included libraries was used for editing the spectra and matching the spectra with the library.

2.4. Contamination measures

Glassware was used for all sampling and digestion steps. All glassware was rinsed with de-ionized (DI) water before use. Digestions were carried out under a laboratory hood and glassware was covered with aluminium foils when no actions were carried out. Lab coats were worn at all times and no synthetic clothes were worn while processing the samples.

Two petri dishes filled with DI water were placed on the hood and at a lab counter every day in order to estimate airborne contamination. Petri dish contents were filtered with DI water and counted to assess contamination levels during laboratory work. Average daily contamination was deducted from overall particle numbers, with respect to days the processing continued for every sample. Filters used were all visually checked under stereo microscope before use.

3. Results and discussions

3.1. Particle colors

A total of 3680 microlitter particles were detected in all of the samples obtained from the WWTP (211 in sludge and 3469 in water samples). Low color diversity was observed in the overall samples investigated where black (57.7%) and blue (28.3%) dominated. Red (8.9%), brown (1.3%), green (1.1%), transparent (1%), orange (0.5%), pink, grey, purple, yellow and white (<0.5%, each) were also observed in all wastewater samples. Particle colors detected in sludge (SLD) were black (56.0%), blue (20.6%), red (7.3%), white (6.0%), green (5.5%), brown (4.1%) and transparent (0.5%). Highest color diversity was observed in the BIO sample. Longer retention times of the aeration basin with slower circulation of particles might have caused such high diversity. Color distributions of wastewater samples for the different steps of the treatment plant and collectively (total wastewater) and for sludge can be found in Figure S2.

Overall, the dominant color observed in samples collected from the different compartments of the WWTP was black. According to the ANOVA test conducted, color distribution differed for the different units of the WWTP ($p < 0.001$) however, it did not change significantly between the grab and 3-hr composite samples ($p = 0.059$). This finding correlates with results of Long et al. (2019) where the dominant color remained the same throughout the WWTP.

3.2. Particle shapes

Throughout the samples, fibers (3407 particles or 92.6%) were the dominant MP type followed by hard fragments (189 particles or 5.2%), soft fragments (64 particles or 1.7%), and others (20 particles or 0.5%). Under the others category, only one piece of a rubber band (most probably) and a pellet were found in the dried sludge sample. The remaining 18 glitter particles under the other category were obtained from the PHC and BIO samples. Several examples of detected particles in the mentioned categories can be seen in Fig. 2.

Particles detected throughout the WWTP are listed in Table S1 according to their sizes and shapes. Whilst hard fragments displayed a contrary behaviour, percent abundances of fibers and soft fragments increased from influent to effluent of the investigated WWTP. Although, the trend was similar in both 3-hr composite and grab samples, their removal rates were different, especially for fibers and soft fragments. Fig. 3 is constructed illustrating shape distributions of particles in all sampling locations and differences of removal dynamics between grab and composite samples. Dynamics of removal was visualised by plotting percent abundance differences between the steps. The graph shows intermittent increase and decreases of the shapes throughout WWTP. Total change of percent abundance was 0% in all sampling locations.

All microplastic particles (<5 mm) could easily pass through the initial bar screens (6 mm) at the entrance of the WWTP. Within the Ambarlı WWTP, 84.1 and 92.3% of fibers were removed from influent to effluent, based on grab and composite samples, respectively. Shares of fibers increased following physical treatment units consisting of screens and the aerated grit chamber. These were detained in biological treatment units consisting of a phosphorus removal unit and aeration basin. In the final settling unit, there was no distinct difference in fiber abundance whereas, less effective removal of fibers was observed for the composite sample.

Fibers have smooth surfaces in general (Anderson et al., 2018) possibly resulting in a reduced resistance to water (Long et al., 2019). Due to their low densities (Andrady, 2017), fibers may escape from the aerated grit chamber rather than settling with the applied current in the wastewater stream. These characteristics could have played a role in their dynamics of detainment in the biological treatment units. Anaerobic and anoxic stages of phosphorus removal might have favoured the settling of fibers with flocs. Furthermore, as biological treatment steps include aeration via diffusors and creation of a bubbled current in the aeration basin, the longer dimensions of fibers could cause entrapment within the foams and aggregates floating in the basin. The agglomeration of fibers, sludge flocs and organic matter occurred in minutes as reported by Schmiedgruber et al. (2019), which indicated reasonable settling of fibers in wastewaters with high organic content. Consequently, some of the fibers should be retained in the sludge. From this perspective, the particles exiting the aeration basin should be further retained in the final settling tank and therefore, their percentage abundance should decrease at least slightly.

High abundances of fibers in the WWTP result from textile laundering. Carney Carney Almroth et al. (2018) observed 400–2478 fiber particles/100 cm^2 released from the washing of textiles with detergent. WWTPs with tertiary treatment units showed a higher presence and lower removal of fibers than secondary WWTPs (Michielssen et al., 2016; Blair et al., 2019), while secondary WWTPs also showed similar characteristics or no significant difference in percent abundance of fibers in some cases (Michielssen et al., 2016; Conley et al., 2019).

Hard fragments were removed from influent to effluent by 94.9 and 97.8%, based on grab and composite samples, respectively. The fragments were mostly trapped within physical treatment units, whereas they slightly increased in biological treatment effluent. Their abundance again slightly decreased following the final clarifier. Hard fragments were retained better with the treatment units applied. Unlike fibers, hard fragments possess lower length to width ratios and angular, twisted, bifurcate, curved and rough surfaces (Helm, 2017). Particles of low density (high buoyancy) or smaller size (low resistance to currents) may have been confined in the aerated grit chamber unit. Biological treatment effluent contained more hard fragments than in the physical treatment unit. Since hard fragments have larger surfaces compared to fibers, they might be more prone to floating in the aeration basin, eventually escaping the basin to the final clarifier. However, they were reported to have captured during secondary treatment processes more

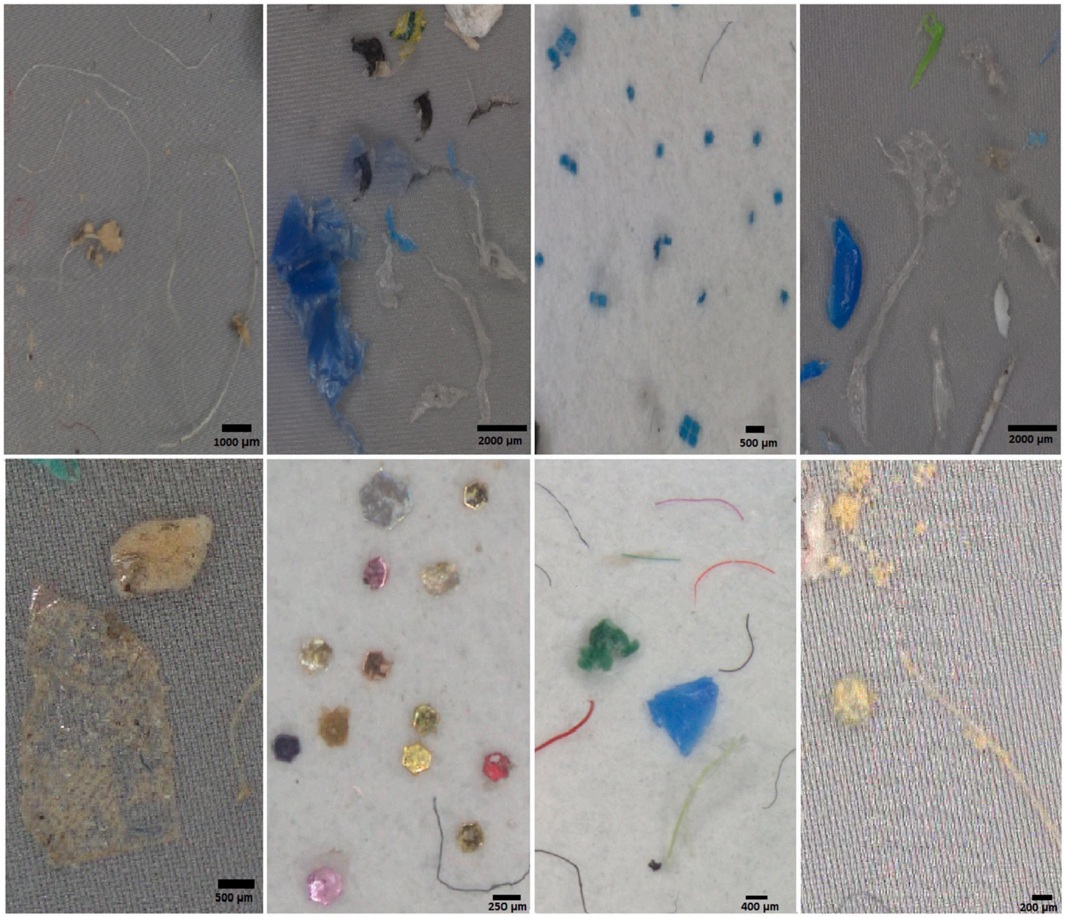

Fig. 2. Different shaped particles extracted from the samples (a-fibers, b-soft fragments, c-hard fragments, d-nylon and several fragments, e-rubber band, f-glitter, g-fragments and fibers, h-pellet and a fiber).

effectively (Long et al., 2019; Liu et al., 2021) except the lamellar structured fragments (Liu et al., 2021). Changes in neutral buoyancies of fragments due to biofilm formation and better removal of fibers with sludge entrapment due to high solids rates can be another reason (Schmiedgruber et al., 2019; Sun et al., 2019). Hard fragments of relatively high density and low buoyancy probably settled independently or with the flocs, which may explain the decrease in percentage abundance of hard fragments in the final effluent.

Soft fragments were generally retained less effectively in all units with the exception of biological treatment with the grab sample. Although this might not be applicable to all particles present in the samples collected in this study, soft fragments are expected to escape the aerated grit chamber since they characteristically display lower length to width ratios compared to fibers but higher than for hard fragments. Therefore, in biological treatment units, soft fragments might act closely to fiber properties due to their elongated shape and they may be entrapped in sludge flocs. In the final clarifier, the soft fragment detention dynamics can be described as between a fiber and a hard fragment. However, many soft fragments escaped the treatment units according to grab samples in the final effluent.

Glitters were observed only in PHG and BIO. However, higher quantities of glitter particles were observed both in counts and percent abundances in BIO, indicating the likelihood of escape from the physical treatment units due to lower buoyancies as glitter particles were absent from the 3-hr composite samples, obtained 1 h before grab sampling took place. Since glitters were observed only in the physical and biological grab samples, a comprehensive evaluation for this category of microplastics could not be made. Glitters have rarely been reported as a distinct group of shapes in studies focusing on WWTPs and are generally reported as hard fragments (Murphy et al., 2016; Yurtsever, 2019). Only one pellet was observed in the sludge. Therefore, no comments can be made on removal of this particle class.

Some WWTPs are reported to remove fibers more efficiently than other particle types, especially hard fragments (Talvitie et al., 2017; Akarsu et al., 2020; Edo et al., 2020). The reverse situation is reported in other studies (Blair et al., 2019; Conley et al., 2019; Magni et al., 2019). In the present study, despite hard fragments being removed more efficiently than fibers, considerably high abundances of fibers was observed within the dried sludge sample. This might be explained by the significantly high abundance of these particles in the influent (Edo et al., 2020).

3.3. Particle sizes

In this study, the majority of particles detected in wastewater samples were in the size range 500–1000 μm, followed by 1000–2000 μm sized particles. In the sludge sample, most particles were of sizes 1000–2000 μm, followed by 500–1000 μm. The overall majority of observed particles fall into the size range 500–1000 μm, followed by 1000–2000 μm and particle sizes increased from influent to effluent (Table S1).

Overall, particles that measured <250 μm and from 250 to 500 μm removed more efficiently than particles >500 μm, especially for hard fragments. However, in the SLD sample, a contrary distribution was observed since particles measuring >500 μm were the most abundant, whereas particles <500 μm only accounted for 13.7% of total particles found in the sludge. Fig. 4 was constructed illustrating size distributions of particles in all sampling locations and removal dynamics based on

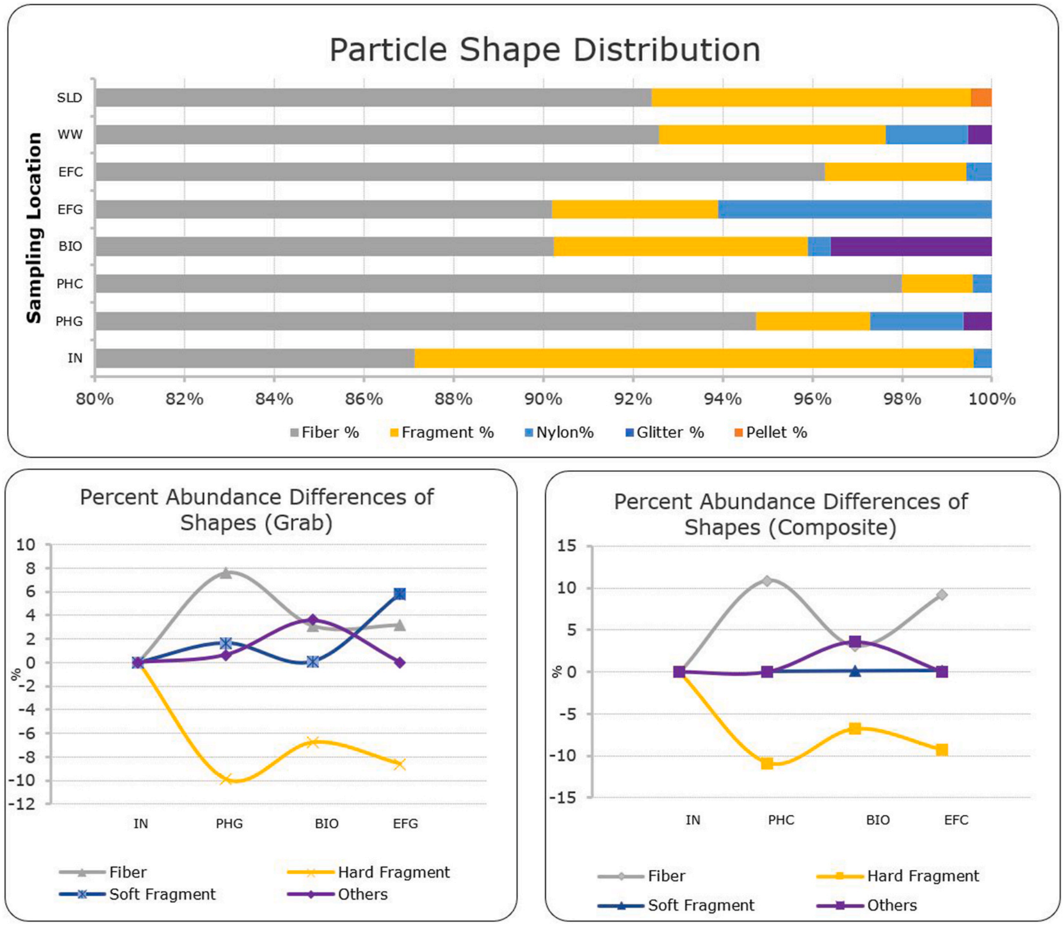

Fig. 3. Frequency distribution of particle shapes and removal rates based on particle shape abundance differences throughout different stages of the WWTP.

percent abundance changes throughout the stages.

Fibers of sizes <1000 μm and >2000 μm, removed more efficiently by the physical treatment units than intermediate sizes. In the biological treatment units, fibers of sizes <500 μm and >2000 μm were removed more effectively than intermediate sized fibers. Fibers in the size ranges 500–1000 μm and 1000–2000 μm displayed contradictory behaviour compared to PHG and PHC samples. Fibers <1000 μm increased while fibers >1000 μm decreased in abundance in both the EFG and EFC samples.

Hard fragments with smaller sizes, especially particles <250 μm were better removed, except for particles measuring >2000 μm in the PHC sample. Notably, hard fragments sized 250–500 μm increased in abundance compared to the PHG sample where they slightly decreased compared to the PHC sample in BIO. In the final settler following BIO, hard fragments of <500 μm decreased in abundance whereas fragments of 1000–2000 μm increased in both the EFC and EFG. Hard fragments sized 500–1000 μm increased in the composite sample of the final effluent but decreased in the grab sample. Hard fragments >2000 μm increased in abundance in the EFG sample however, no change was observed for the EFC sample.

Soft fragments with different sizes displayed contrary behaviour throughout WWTP units, such as increase in soft fragments was observed in the EFG sample where no change was detected for the EFC sample. As soft fragments did not show similar patterns for grab and composite samples throughout the WWTP units, their removal dynamics could not be commented on. Glitter particles were also more abundant in BIO compared to PHG, but was absent in the subsequent phases.

Overall, despite the differences in removal efficiencies between composite and grab final effluent samples and the raw influent sample, abundance of fibers of sizes <250 μm and 500–1000 μm increased whereas 250–500 μm and 1000–2000 μm size ranged fibers decreased. Hard fragments measuring <500 μm were removed almost completely from the WWTP units whereas the remaining microplastic types generally increased in abundance. Several soft fragment size categories seen in the raw influent sample could not be detected in the composite final effluent sample, however, average sizes decreased. Glitters were only detected in the PHG and BIO stages with increased abundance in BIO.

The sludge sample contained the most fibers in the size range 1000–2000 μm and fibers >2000 μm remained slightly increased or decreased compared to the aforementioned sizes in composite and grab samples, respectively. Hard fragments displayed the most significant decreases from influent to effluent and were either found in low numbers or totally absent in the sludge sample, where fibers that were increased in the final effluent sample were the most abundant. Soft fragments or glitter were absent in the dried sludge sample. Only one pellet was detected in the sludge, where no pellets observed in the wastewater samples.

Most studies in the literature, regarding the dynamics of removal agree with our findings, for particles <500 μm and particularly 250–500 μm size particles (Li et al., 2018; Conley et al., 2019). Stepwise removal showed minor differences resulting in higher or lower sizes in different units, or better/worse removal of different size classes for different shapes (Bayo et al., 2020). Overall, considering the particle shape and size classes in particle removal dynamics, those findings indicate removal of smaller hard fragments and small to mid-sized fibers

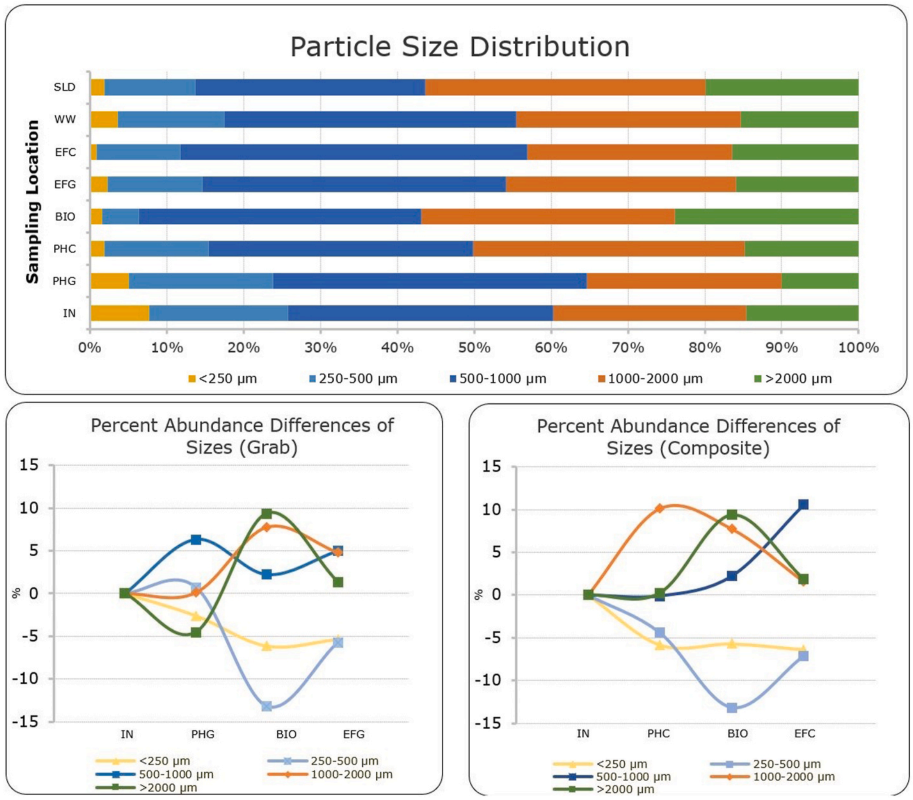

Fig. 4. Frequency distribution of particle sizes and removal rates based on particle size abundance differences throughout different stages of the WWTP.

throughout the treatment phases to be more efficient than for other size classes. Mid-sized fibers were not removed as efficiently and this might be caused from more successful removal of smaller particles in aerated grit chamber units and better entrapment of larger fibers in sludge flocs or settling in final clarifiers. Soft fragments abundance affected the removal rates of different size classes more than other shapes (Gündoğdu et al., 2018). As soft fragments were less common than hard fragments in Ambarlı WWTP, their influence on size averages and removal dynamics was not relevant compared to hard fragments and especially fibers.

In some cases, larger differences in removal dynamics were observed compared with our findings despite the general dynamics being similar (Edo et al., 2020). Different removal dynamics were observed where the influent abundances of smaller particles were higher or abundances of shapes were different with similar size distributions (Talvitie et al., 2017; Magni et al., 2019; Park et al., 2020). Lares et al. (2018) reported a similar influent distribution to Ambarlı WWTP however, particles of sizes <250 µm and >1000 µm were retained less and particles sized 250–500 µm retained more than the remaining particles. These differences resulted in larger average sizes of particles in Ambarlı WWTP.

In summary, the removal of particles depends on particle size and shape. The overall removal dynamics for all investigated parameters combined were similar to most studies conducted on tertiary WWTPs and some secondary WWTPs with advanced nutrient removal steps. The results mostly contradicted the studies with lower mesh sizes. Parameters such as operating conditions, wastewater characteristics can also affect the removal dynamics.

3.4. Average sizes

Average sizes were calculated for samples collected from the WWTP. Average length was determined as 1223 µm for all particles detected, where maximum length was 5.00 mm and minimum length was 0.05 mm for all particles detected. Average size increased from influent to effluent of the WWTP by nearly 9%, due to larger contribution of fibers with sizes 500–1000 µm and >2000 µm. Akarsu et al. (2020) similarly reported that overall average microplastic length was surprisingly higher in effluent waters (1309 µm) compared to influent (1135 µm). These authors suggested that such observation could be due to displacement of some of the microplastics retained for longer times in the WWTP. Average sizes for all sampling locations and for all shape categories are given in Figure S3.

The average particle sizes observed at Ambarlı WWTP (Istanbul) were similar to the study conducted by Yang et al. (2019) which used a 50 µm mesh used for particle collection. Karaduvar WWTP (Mersin), which applies tertiary treatment for phosphorus removal, showed higher average particle sizes despite a smaller (26 µm) mesh being used for particle filtration (Akarsu et al., 2020). In the latter study a higher rate of size increase from influent to effluent was reported, however, there was no clear correlation with particle shapes and differences observed with average sizes. From particle shape distributions reported in several studies, it can be generally concluded that with higher abundances of fibers and soft plastics, a higher average length value is observed. On the contrary, a higher presence of hard fragments correlated with smaller sizes, regardless of the removal dynamics that differed throughout WWTPs reviewed (Akarsu et al., 2020; Edo et al., 2020; Gündoğdu et al., 2018; Mason et al., 2016).

Samples collected from Ambarlı WWTP had a high organic content in all samples, especially from the biological treatment units. Higher suspended solid concentrations correlated with greater abundances of MPs >1000 µm, whereas lower suspended solid quantities corresponded to MPs <1000 µm according to Bayo et al. (2020). Therefore, this factor might also have contributed to the higher average sizes observed here compared to results reported in other similar studies. Daily fluctuations in concentrations was confirmed by Talvitie et al. (2017), indicating that daytime microplastic concentrations were higher than for night-time.

This finding was also supported by Bayo et al. (2020) who stated differences in the morning size average size (660 μm) compared to the afternoon average size (790 μm). As the samples from Ambarlı WWTP were collected in the afternoon, increase in size averages could have occurred from the time of sampling.

3.5. Chemical characterization

Particles extracted from the influent sample, grab effluent sample and sludge sample were analyzed with a Raman microscope. 10 particles randomly selected from each sample category were analyzed. Following removal of spikes, baseline corrections were conducted for the original spectral data and compared with spectra from library reference records. 10 particles, 7 particles and 7 particles yielded matching results to spectra with a ratio above 60% from the particles in the influent sample, effluent sample and sludge sample, respectively. Only 2 particles were characterized as plastics (Polycarbonate (PC) and Polyurethane (PUR)) in the influent sample. 3 particles were identified as plastics in the effluent sample (PC particle, PUR foam and Polyethersulfone (PES) film with epoxy coating) with a high matching score with one possible composite containing 12% PC or PUR in its structure. In the sludge sample, 4 particles were characterized as plastics (PC, Polyethylene terephthalate (PET), PES and one categorized as PES or PET). Particles identified as non-plastics were salts, rutiles and other organic matter of natural origin and graphite, pharmaceuticals and colorants of unnatural origin. Examples of plastic and non-plastic spectra with matching reference spectra are given in Figure S4-S9.

In total, 16 of 30 particles (53%) extracted from several phases of the WWTP were confirmed as plastics. Confirmation ratios of particles samples are dependent on studies in the literature and range from 40% to 75% according to the number of particles analyzed in the study and size ranges investigated (Edo et al., 2020; Lenz et al., 2015). The ratio found here of 53% therefore was acceptable. Moreover, the spectra quality was low and colorants, pharmaceuticals and carbon readings may also represent materials adhered to the surface of plastics. The particles subjected to characterization were also low in quantity, therefore, the plastics confirmation ratio could have been underestimated.

3.6. Removal and discharge of particles

In total, 137.0 ML/L entered Ambarlı WWTP, where 21.1 ML/L, 9.7 ML/L and 60.3 ML/g exited the WWTP with the EFG, EFC and SLD samples, respectively. Particle concentrations and removal rates are given in Table 1 and Table S2, respectively. Considering the plastics confirmation rate for overall particles characterized, the influent microplastic (MP) concentration was calculated as 72.6 MP/L. Effluent particle concentration ranged from 11.2 to 5.1 MP/L and the average effluent concentration was 8.2 MP/L. Sludge particle concentration was 32 MP/g.

Calculated from the average particle concentrations in EFG and EFC samples and flowrate of the WWTP, an average of 5,545 $\times$ 10^6 ML/day (2,934 $\times$ 10^6 MP/d) were released in the receiving water channel. The WWTP also produced dried sludge corresponding to 6,071 $\times$ 10^6 ML/day (3,218 $\times$ 10^6 MP) for final disposal.

Table 1
Microlitter and microplastics concentrations (particles per liter) in the samples after corrections for contamination.

Particles	IN	PHG	PHC	BIO	EFG	EFC	SLD (Particles/g)
Fiber	119	54.6	63.3	344	18.8	9.2	55
Hard fragment	18	1.6	1.1	22	0.9	0.4	5
Soft fragment	1	1.3	0.3	2	1.4	0.1	0
Glitter	0	0.4	0	14	0	0	0
Pellet	0	0	0	0	0	0	0.3
Total (ML)	138	57.9	64.7	382	21.1	9.7	60.3
Total (MP)	73.1	30.7	34.3	203	11.2	5.1	32

Most of the WWTPs reviewed in the literature reported lower effluent concentrations of <2 MP/L (Murphy et al., 2016; Lares et al., 2018; Li et al., 2018; Blair et al., 2019; Long et al., 2019; Magni et al., 2019; Park et al., 2020; Bayo et al., 2020), compared to an average of 8.2 MP/L found in this study, as their influent concentrations were lower, regardless of the removal rates. Higher concentrations (56–65 ML/L) were reported for several WWTPs reviewed by Leslie et al. (2017), where no characterization was applied. Particle concentrations in the effluent were similar (10–30 MP/L) to some studies with different WWTPs investigated (Liu et al., 2019; Edo et al., 2020). Conley et al. (2019) reported concentrations ranging from 1 to 30 ML/L, which also showed a similar trend of removal with Ambarlı WWTP, with no characterization step applied. Concentrations of MP (10.7 MP/L) and ML (26 ML/L) were reported by Edo et al. (2020), where ML concentration following biological treatment was also similar (451 ML/L) to this study. Removal rates did not change significantly with microlitter and microplastics in the study.

Overall removal rate for Ambarlı WWTP (84.7–93.0%) was generally in line with other European WWTPs, where 72–99.9% removal was observed. Particle removal rates were better than for other facilities reported in Turkey, where 73–79% removal of particles in WWTPs in Adana (Gündoğdu et al., 2018), 38–78% removal of particles in WWTPs in Mersin (Akarsu et al., 2020) and 79.5% removal of particles in a WWTP in Sakarya (Bilgin et al., 2020) were reported. Concentrations observed in effluent samples varied between 0.6 and 7.2 MP/L, which were again lower than those of Ambarlı WWTP in average, regarding the ML and MP values reported in the aforementioned studies. These differences can be explained by the treatment plants receiving lower concentrations of particles according to the data presented in the studies.

Findings from Ambarlı WWTP also confirmed that considerable amounts of particles were removed during physical treatment steps as stated in previous studies (Murphy et al., 2016; Michielssen et al., 2016; Lares et al., 2018; Yang et al., 2019; Magni et al., 2019). Results obtained in our study were also in line with studies stating that WWTPs with tertiary treatment and secondary WWTPs with phosphorus/nutrient removal units removed MPs more efficiently (Park et al., 2020).

Discharged particles were significantly higher than 100 and 133 MP/capita/day that were reported by Murphy et al. (2016) and Magni et al. (2019), respectively as the plant's release was calculated as 1467 MP/capita.day based on nearly 2 million people that the WWTP serves. Studies conducted in the Sea of Marmara investigating microplastic concentrations reported a range of 0.012–47 MP/L where higher values were obtained from a study conducted in a coastal region affected by discharged wastewater and incoming currents (Faruk Çullu et al., 2021). Tunçer et al. (2018) observed more uniform and lower values in samples obtained from estuaries, open sea samples and straits (Tunçer et al., 2018). Considering both of these studies, the concentrations of effluent discharge (5.1–11.2 MP/L) is still considerably significant compared to the environment that it is discharged into. This range of value corresponded to a concentration, 670-fold of offshore concentrations of MPs and 20% of coastal MPs samples from an estuary observed in Marmara Sea in the aforementioned studies.

Sludge produced at Ambarlı WWTP contained 1600–5,640 $\times$ 10^6 MP/day in line with quantities found in dry sludge samples reported by Li et al. (2018). Particle concentrations in sludge were also affected by influent concentrations as well as the effluent sample concentrations (Murphy et al., 2016; Leslie et al., 2017; Mintenig et al., 2017; Lares et al., 2018; Talvitie et al., 2017). Sludge particle concentrations were generally found to be higher than influent concentrations excepting studies by Edo et al. (2020) and Lee and Kim (2018). Dominant particles in the influent sample also occurred in the sludge samples and were considerably high in both (Lee and Kim, 2018; Edo et al., 2020).

3.7. Other parameters

Most of the studies reviewed in the literature were conducted in secondary WWTPs with nutrient removal units and tertiary WWTPs as mentioned above. Regarding the similarities of the treatment processes utilized, removal dynamics should have been similar with our findings. Differences in facilities with similar treatment units and influent distributions suggests that WWTP design and operation can be another factor affecting particle removal. (Gündoğdu et al., 2018; Lares et al., 2018; Long et al., 2019; Park et al., 2020). Removal dynamics can also be affected by suspended solids content as mentioned above, pH levels and humic acids presence (Li et al., 2018; Bayo et al., 2020). Incoming particle sizes and shapes could also have influenced the removal dynamics as mentioned in previous sections. These combined parameters may affect removal of particles which did not always correlate with abundance of particle shapes (Akarsu et al., 2020).

Wastewater load and characteristics change seasonally, monthly and even daily (Talvitie et al., 2017; Conley et al., 2019). As reported by Akarsu et al. (2020), diversity of particles observed in influent streams and efficiency of the WWTP at retaining different types of particles can change throughout the year. Karaduvar WWTP is a good example for both such cases, whereas Tarsus and Silifke WWTPs illustrate how the plant responds differently to even minor fluctuations in influent characteristics, during different times of the year (Akarsu et al., 2020). These findings further indicate variation in wastewater content and operations affecting the removal performance of WWTPs.

4. Conclusions

The findings of this study showed that the WWTP under investigation removes the bulk of microlitter and microplastics from sewage however, loads from the WWTP effluent were comparably higher than shoreline and open sea samples, confirming that WWTPs constitute a considerable source of microplastics contamination to the Sea of Marmara. Removal dynamics were observed to be similar for both MPs and ML, for the size categories that were abundant in our study, where particles <250 μm were better removed in all particle shape classes. Particle sizes and morphologies played an important role in dynamics of removal. However, external factors and operation conditions as well as dynamics of removal for particles <25–50 μm, could have caused discrepancies between similar WWTPs reviewed in literature and should be further reviewed.

WWTPs are both recipients and sources of microplastics since they receive high loads of microplastics, but are unable to retain all particles with currently applied treatment technologies. Therefore, use of different units such as coagulation vs. floatation towards a better removal efficiency as well as possible additions or modifications of treatment units with emphasis on MP and ML removal which facilitates better entrapment of the particles in the sludge are important. However, more importantly, reducing the loads of microplastics to the WWTPs should be addressed in the first place, in order to reduce emissions via effluent streams of these facilities. Loads of MPs into WWTPs should be reduced by approaches such as restricted use or ban of products containing primary MPs and other products such as ear buds that are made of plastics or sanitary wet towels that increase the plastic loads to WWTPs. Legislative implementations and social awareness are crucial to facilitate this change.

Suat Vardar: Conceptualization, Methodology, Investigation, Writing - Original Draft Turgut T. Onay: Supervision, Writing - Review & Editing. Burak Demirel: Funding acquisition, Writing - Review & Editing. Ahmet Kıdeyş: Investigation, Writing - Review & Editing.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2021.117862.

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