Microplastic pollution in wastewater treatment plants in the city of Cádiz: Abundance, removal efficiency and presence in receiving water body

Franco, A. A. [a]; Arellano, J. M[b*]; Albendín, G. [b]; Rodríguez-Barroso,R[a]; Quiroga, J. M[a] [a]; Coello, M[a].D.[a].*

[a]*Environmental Technologies Department, University Institute of Marine Research (INMAR), International Campus of Excellence of the Sea (CEI MAR), Faculty of Marine and Environmental Sciences, University of Cadiz, 11510 Puerto Real, Cadiz, Spain*

[b] *Toxicology Department, University Institute of Marine Research (INMAR), International Campus of Excellence of the Sea (CEI MAR), Faculty of Marine and Environmental Sciences, University of Cadiz, 11510 Puerto Real, Cadiz, Spain*

Corresponding author: juanamaria.arellano@uca.es/ anafrancodelpino@gmail.com

Keywords: *Wastewater treatment plant, Microplastics, FTIR, receiving water body, Polymers.*

Abbreviations

ASA	Acrylonitrile styrene acrylate
DAP	Diallyl Phthalate
CA	Cellulose acetate
EAA	Ethylene acrylic acid
EVA	Ethylene-vinyl acetate
HDPE	High density polyethene
MP	Microplastics
PA	Polyamide
PAH	Polycyclic Aromatic Hydrocarbon
PB	Polybutylene
PCB	Polychlorinated biphenyl
PCL	Polycaprolactone

PE	Polyethylene
PET	Polyethylene terephthalate
PMMA	Poly(methyl methacrylate)
PP	Polypropylene
PR	Phenoxy resin
PS	Polystyrene
PVC	Polyvinyl chloride
WWTP	Wastewater treatment plant

Abstract

Wastewater treatment plants (WWTPs) have been appointed as one of the main sources of microplastics (MP) into marine ecosystems. The aim of this research work has been to study the influent and effluent of two WWTPs, both located in Cádiz, with different wastewater source (industrial and urban), as well as the receiving water bodies where the facilities discharge their sewage.

MP were collected, extracted from wastewater matrixes and analysed according to the abundance, shape, size, and type of polymer, along with the removal rates of MPs in the plants.

Subsequently, the data obtained on both WWTPs were compared, the main difference among the WWTPs was the amount of microplastics found in the wastewater, as well as the presence of polymers with resins from industrial activities.

The results from this study established that the most representative form was fibers; about the size, 100-355 μm fraction was the most abundant, followed by 355-1000 μm and finally the size among 1000-5000 μm. Regarding to the type of polymers, 17 were identified using attenuated total refraction Fourier-transformed infrared spectroscopy (ATR-FTIR). Further, PVC, PE, EAA and HDPE were the largest found polymers.

The presence of MPs in the influent varied from 645.03 ± 182.24 MPs/L to 1567.49 ± 413.18 MPs/L in the urban and industrial WWTP respectively; in the effluent, it varied from 16.40 ± 7.85 MPs/L to 131.35 ± 95.36 MPs/L. The removal rate overcome the 90% in all the samples.

Receiving water bodies presented heterogeneous abundance of microplastics 6.64 ± 2.71 MPs/L and 0.83 ± 0.26 MPs/L in the zones close to IWWTP and UWWTP discharge point.

The results obtained shows that despite the elimination efficiency in the WWTPs studied, these facilities act as a significant source of MPs into aquatic ecosystem due to large flow of water discharged.

1 Introduction

Microplastics (MP) are plastic particles smaller than 5 mm (Gago et al., 2016; Sun et al., 2019; Talvitie et al., 2017; Thompson et al., 2004). These emerging contaminants have sparked interest in news media, education institutions, and society because they have been detected ubiquitously in animals, soils, and water bodies (freshwater, brackish, and marine) generating widespread alarm (Anderson et al., 2016; Asensio-montesinos et al., 2020; Conley et al., 2019; Hann et al., 2018; Li et al., 2015). The plastic global industry produced 360 million tonnes in 2018, of which 29 million tonnes were recycled and treated (Plastics Europe., 2019); so presumably around 90% of the plastic produced were untreated waste that may reach the natural environment, degrading and contaminating the aquatic system due to the durability and resilience of plastic.

MPs can be classified as primary or secondary. Primary MPs are manufactured in sizes smaller than 5 mm and widely used in cosmetics, but also in hygiene products, detergents, and fibers released from laundry (Napper et al., 2015; Cristaldi et al., 2020; Sol et al., 2020; Bretas Alvim et al., 2020). Secondary MPs become micro in size through physical, chemical, and/or biological degradation processes of larger plastic (Gatidou et al., 2019; Sun et al., 2019).

In recent years, studies of MPs in the marine environment have been conducted to detect the presence, interaction, and deposition of MPs in water bodies, fauna, sediments, and saltworks (Browne et al., 2011; de Sá et al., 2018; Iñiguez et al., 2017; Long et al., 2019; Nel and Froneman., 2015). The results of these studies demonstrate that these pollutants pose a threat to the ecosystem and organisms that inhabit it because MPs can absorb other pollutants (such as PAHs and PCBs) enhancing their contamination (Alimi et al., 2018).

In Spain, there are any policies to decrease the amount, production, and release of MPs; however, few EU member states (France, Italy, Sweden) have introduced bans or restrictions on the use of tiny plastic spheres in personal hygiene products. In addition, EU Regulation 2020/741 established requirements for reuse of water and states that MPs and micropollutants should be studied to protect the environment and living organisms (Franco et al., 2020; Vuola et al., 2019).

Microplastics can reach the marine environment through multiple pathways, such as poorly managed landfills, stormwater runoff, windborne waste, untreated sewage, and offshore activities (Hann et al., 2018; Sundt et al., 2016). Effluents from wastewater treatment plants (WWTPs) are another important route for MP to enter the aquatic environment (Sun et al., 2019; Talvitie et al., 2015). These facilities receive and treat wastewater from domestic, urban, and industrial activities to avoid contamination when the water is returned to the environment or reused. WWTPs were not designed to remove microplastics from wastewater, however, removal efficiency can range from 64 to 99%, and sludge is expected to be the final fate of MPs retained during depuration at a conventional WWTP (Elkhatib and Oyanedel-Craver, 2020; Habib et al., 2020; Sol et al., 2020). Despite the high removal efficiency, it is estimated in the order of 10^9 MPs can be released into the environment daily (Hidayaturrahman and Lee, 2019).

The present study focused on MP contamination in WWTPs in the city of Cádiz (southwest of Spain)—one industrial and one urban—and the presence of these pollutants in the receiving water to determine the abundance of MPs in sewage samples from these WWTPs with respect to their shape, size, and polymer type; calculate the removal efficiency of the facilities; and estimate the amount of MPs released into the environment.

2 Materials and Methods

2.1 WWTP samples

Wastewater samples were collected from two WWTPs in the city of Cádiz, Spain in 2019. Different treatment capacity, population equivalent, influx composition, and water treatment at both facilities were compared (Table 1). The WWTPs analysed are the only ones located within Cádiz's city limit (Figure 1). The urban WWTP of Cádiz had a treatment capacity over 19 million m^3/year serving the inhabitants of Cádiz and San Fernando city in which effluent is discharged into the sea through an underwater outfall.

The industrial WWTP was designed to treat 30,000 m^3/year of sewage from vessels and ship building and reparation; after depuration, the water is dumped directly into the port of Cádiz. Both WWTPs discharge their effluents into the Atlantic Ocean. To study the presence of MPs in the receptor water body, samples were collected from two zones (Figure 1).

2.2 Microplastic sampling

Sampling at both WWTPs and in the receiving waterbodies was conducted in spring 2019. Influent sewage samples were collected in the influent after passage through perforated screens and before the mixing of wastewater with the recirculated sludge. Effluent samples were taken prior to discharge points after disinfection (Masura et al., 2015; Xu et al., 2019); however, sampling points had to be adapted in the effluent of the industrial WWTP, due to difficulty in sampling conditions. Wastewater samples were collected using a steel scuttle, then filtered through stainless steel sieves of various mesh sizes (1000, 355, and 100 µm). Heterogeneous sewage composition, population habits, and variations in sewers systems hinder the ability to measure the volume of wastewater sampled; the volume of influent collected varied from 3–10 L, whereas the volume of effluent sampled ranged of 15–35 L. Particles retained on the stainless steel sieves were transferred into beakers using distilled water and letting them dry.

2.3 Sample extraction

Wastewater contains a complex matrix with digested labile matter that needs to be removed. In the present study, the wet peroxide oxidation (WPO) method was used (Magni et al., 2019; Masura et al., 2015; Ou and Zeng., 2018; Xu et al., 2019). This procedure was recommended by National Oceanic and Atmospheric Administration (NOAA) based on the addition of 20 mL of aqueous 0.05 M Fe (II) solution and 20 mL of 30% hydrogen peroxide (H_2O_2) into the beakers containing the samples. Subsequently, a magnetic stir bar was added, and the samples were stirred at 75 °C and 90 rpm for 30 min. After exothermic reactions, samples were transferred to a separating funnel to sort the particles by density. Finally, the samples were filtered through a glass sand core filter and placed in polycarbonate filters.

In the case of seawater samples, no extraction method was needed. The samples were filtered through stainless stell sieves, transferred to beakers using distilled water, filtered through a glass sand core filter, and placed in polycarbonate filters.

2.4 Sample characterization

After organic digestion, MPs were distinguished according to their morphological and chemical characteristics.

2.4.1. Morphological characterization

Physical analysis of samples was based on visual examination, counting, and classifying the MPs according to morphological characteristics of size and shape using a Carl Zeiss Axio Imager M1m optical microscope. Samples were distinguished in five shapes (fibers, spheres, filaments, flakes, and fragments). Visual identification is prone to miscalculation due to the complexity of discriminating the particles, which can lead to underestimation or overestimation of particle abundance (Franco et al., 2020; Iyare et al., 2020; Masura et al., 2015; Sun et al., 2019).

2.4.2 Chemical characterization

Chemical characterization was based on spectroscopic methods used to identify the types of polymers in the samples collected using a PerkinElmer Spectrum 100 Fourier transform infrared spectroscopy (FT-IR). To determine the composition of the MPs, particles were exposed to infrared radiation (Sun et al., 2019), generating a specific spectrum for each particle depending on the chemical bonds between the atoms. The outcome spectrum was analysed using characteristics peaks compared to the polymer library of peaks in the reference spectrum (Gago et al., 2016; Ou and Zeng, 2018; Torre, 2015).

2.5 Contamination control

To prevent contamination, all materials used were cleaned with alcohol and plastic lab ware were avoided during this study. All samples were covered using watch glass; lab coats and gloves were worn during all procedures, and a blank filter was exposed to the air during sample characterization of each sampling point.

2.6 Statistical analysis

The concentrations of MPs were calculated considering the total amount of MPs and the volume sampled (Equation 1). Results were presented as the mean ± standard error in units of MP/L.

$$MP\ Concentration = \frac{Number\ of\ MPs}{Volume\ sampled\ (L)}\ . \qquad (1)$$

Removal efficiency (RE) was estimated considering the concentration of MPs in the influent and effluent (Equation 2):

$$RE = \frac{MP\ concentration\ influent - MP\ concentration\ effluent}{MP\ concentration\ influent} \times 100\%\ . \qquad (2)$$

3 Results and Discussion

3.1 Microplastic occurrence and removal efficiency

Not all particles collected in samples were plastics (Gies et al., 2018). Figure 2 shows MP proportions relative to total microparticles found at each facility and sample point. Non-MP particles were identified as additives, plasters, hormones, cellulose, or polymers; if the search coincidence was below 70%, the particles were not considered to be MP (Franco et al., 2020; Frias et al., 2020).

Microplastics were widely detected at both facilities (Table 2). The concentration in the urban WWTP was 645 MP/L in the influent and 16 MP/L in the effluent; whereas the abundance was greater in the industrial WWTP, up to 1567 MP/L and 131 MP/L in the influent and effluent, respectively. These results are consistent with other studies of MPs in urban WWTPs (Franco et al., 2020; Sun et al., 2019; Magni et al., 2019) No specific studies on the presence of MPs in the industrial WWTP were found; but a large gap was found between the concentration of MPs in the urban and industrial WWTPs analysed in the present study. This variation could be explained by the source, and use of water; the urban WWTP serviced a major population (Cádiz and San Fernando cities) and received wastewater from residential and domestic activity, while the industrial WWTP treated sewage from building, cleaning, and repairing of vessels and ships; these activities require large amounts of paint, coating, anti-skid powder, and abrasive materials composed of synthetic polymers which may contribute to the higher concentration of MPs in the industrial facility.

The RE were calculated for both WWTPs, and the urban facility presented a 97.46% MP removal rate, while the industrial WWTP removed 91.62% MPs from the water line during depuration. These results are consistent with previous studies on MP RE in WWTPs (Table 2) (Edo et al., 2019; Lares, 2019; Murphy et al., 2016; Sun et al., 2019). However, comparison of RE in different studies is subject to inaccuracy due to the large and heterogeneous range of MP concentrations, and the lack of standardized methods of sampling, treatment, and quantification makes comparisons challenging across the consulted research (Gatidou et al., 2019; Ziajahromi et al., 2017).

Despite the high removal rate, a daily average of $1.49–1.94 \times 10^9$ MPs/day were discharged into the Atlantic ocean from the urban WWTP, whereas $1.07–2.64 \times 10^7$ MPs/day were discharged into the ocean from the industrial WWTP during the studied period, however it is important to prolong the investigation to determine Microplastics release fluctuation for a longer time period. Although the industrial WWTP had more

MPs concentration than the urban WWTP, the minor daily flux in the industrial facility means fewer MPs enter the environment. Nevertheless, the amount of MPs discharged into the marine environment is significant, which confirms that WWTPs are conduits of MPs to the environment.

3.2 Size and shape of microplastics

Size and shape are physical characteristics studied of microplastics because they impact the capacity of depuration to remove these particles from the sewage during treatment. In addition, these features affect adhesion of other pollutants, plasticizers, and microorganisms (Iyare et al., 2020; Liu et al., 2019).

With respect to size, particles under 355 μm comprised over 50% of the total MPs in each sample (Figure 3.A) in both influent and effluent; thus, no notable significant difference was detected between them. The comprised more than 70% of each sample, which is consistent with previous studies (Conley et al., 2019; Edo et al., 2019; Hidayaturrahman and Lee., 2019; Sun et al., 2019; Xu et al., 2019). The greater abundance of smaller particles, rather than larger is attributed to fragmentation of larger plastics during transport to and through the sewer system or the retention of bigger MPs throughout the treatment process. Simon et al. (2018) proposed that physical retainment by sedimentation is the principal removal mechanism for most MPs at the WWWTP.

Figure 4 shows an example of each shape founded in the present study. With respect to shape distribution, fibers were the most abundant shape representing over the 40% of all the particles in all of the samples from both facilities, followed by fragments and flakes; films and spheres were less common shapes (Figure 3.B). In other studies, fibers were also the predominant shape (Franco et al., 2020; Gies et al., 2018; Iyare et al., 2020), and it is attributed to the release of plastic fibers during laundry process. Salvador et al. (2017) reported that a single piece of clothing can release up to 1,900 fibers in a single wash. On average, a regular 6 kg domestic washing machine can discharge 700,000 fibers into the sewage system during laundering (Napper & and Thompson, 2016). Fibers are difficult to retain during depuration due to their shape (long and narrow) which inhibits their retention in conventional WWTPs (Sun et al., 2019).

Fragmentation of large plastic items during usage, cleaning, and maintenance has been proposed as the origin of plastic fragments and flakes (Sun et al., 2019; Xu et al., 2019) characterised by irregular and rounded shapes, respectively. Similarly, films and spheres were not common shapes found in previous studies as well, with a concentration below 10% (Talvitie et al., 2015; Xu et al., 2019). In the case of spheres, these particles are used in cosmetics (toothpaste, exfoliants, and soaps), but their use has been banned in some European countries causing manufacturers to stop including MPs on their products, resulted in a decrease of spheres in wastewater in recent studies (Edo et al., 2019; Napper et al., 2015; Sundt et al., 2016).

3.3 Polymer identification

The FT-IR spectroscopy revealed 14 different polymers in the samples (Figure 5). The most common types of polymers were PVC, HDPE, PE, and EAA found in most of the samples. These four types of polymers are among the 10 most-demanded and manufactured plastics in the world (PlasticsEurope, 2019), which explain their abundance in the WWTPs analysed in this study; these polymers were also the most abundant in other studies (Liu et al., 2019; Xu et al., 2019). These polymers are thermoplastics widely used to manufacture plastic containers, bottles, pipes, clothes, facemasks, toys, tool coatings, paints, cable and wire sheathing, and so on, explaining their high presence in both urban and industrial wastewater.

Regarding the urban WWTP, PA was identified in influent and effluent, this polymer is formed by synthetic fibers used in clothing and toothbrushes, which can be released during laundering and personal grooming. Despite the higher percentage distribution of MPs in effluent (40%) related to influent (5%), the concentration (MP/L) in the influent of 32.25 MP/L is larger than the concentration in effluent (6.56 MP/L). Table 3 shows concentrations (MP/L) according to polymer type in the present study. PMMA was identified in the influent of both facilities, this polymer is used for the manufacture of products as diverse as contact lens and transport covers in industry. EVA and PP were also found in the influent at the urban WWTP. These plastics are used in households for domestic and recreational activities such as food packaging, wrappers, and crafts.

Regarding effluent from the urban WWTP, four polymers were found: HDPE, PVC, PA, and PS. It should be noted that PS was not identified in the influent samples; this might be due to the heterogeneous composition of the sewage or the use of this polymer as an insulator in the facility that releases these particles into the treated water. In the industrial WWTP, the most abundant and demanded plastics mentioned before were present at both sample points (influent and effluent). The polymers PMMA, PS, PET, and PB were also identified in the influent.

With respect to the effluent in the industrial WWTP, eight polymers were found. HDPE, PE, EAA, and PVC were the most abundant (above 10% each). Less common plastics were ASA, DAP, PP, and PCL, which are stable, flame retardant, and resistant to oil, fuel, and solvents, characteristics contribute to the presence of these polymers possible in industrial wastewater. Our results showed great heterogeneity in the nature of the MPs from two types of treatment plants, one industrial and the other urban. Therefore, a more exhaustive study is essential, increasing the number and type of treatment plants to be sampled, with the aim of knowing in greater depth the behaviour and nature of the MPs discharged into the environment.

3.4 Microplastics in receiving water

Receiving water exhibited heterogeneous abundance of microplastics. In the case of zone 1, influenced by the urban WWTP, it was concluded that an average of 0.83 ± 0.26 MP/L was present in the water; whereas zone 2, within the discharge point of the industrial WWTP, the concentration of MPs was 6.64 ± 2.71 MP/L. These results are consistent with previous works; for example, Zhang et al. (2018) reported 0.74 MP/L in the Bay of China. Considering previous results obtained by Ng and Obbard (2016) and Nel and Froneman (2015), the amount of MPs found in the Bay of Cádiz was higher than those found in the waters of Singapore and South Africa, respectively. Nevertheless, the differences observed in MP content is not entirely conclusive because the treatment of samples were not standardized.

Figure 6.A shows the difference in MP content observed at the two sampling points. Zone 2, close to the Port of Cádiz, presented a higher load of microplastics in comparison to zone 1. This is probably because most of the particles found might come from industrial activities that take place in the area adjacent to the discharge of the industrial WWTP, within the port of Cádiz (Zone 2). For this reason, it is not possible to ensure that the particles observed in the sample from zone 2 originated in the effluent of the industrial WWTP. On the other hand, the concentration of MPs found in zone 1 was low, although it was above values observed in coastal areas not affected by WWTP discharges.

The shapes of the MPs in zones 1 and 2 provide useful information about their source (Figure 6.B). Microparticles in zone 1 were predominantly fibers, as described by other authors (Salvador et al., 2017; Wagner et al., 2018). On the other hand, fragments were predominant in zone 2, indicating a strong influence from the nearby industrial area. The difference in the shapes of particles found in the samples was probably motivated by the high heterogeneity of the water bodies under study. Liu et al. (2019) and De Sá et al. (2018) detected that the predominant forms were fibers, except for one sampling point where the predominant form was fragment, which corroborates the distribution observed in this study.

As above-mentioned, zone 2 is in a port area with continuous maritime traffic and therefore expected to discharge more than that the amount found in marine areas with less human activity (Zone 1) (Norén, 2007).

Figure 7 shows the different polymers (mean values) determined in each sample. PE was identified in all of the samples. HDPE and PA were only found in zone 2. In zone 1, only three polymers were found: CA (40 %), PA (20 %), and PE (40%).

4 Conclusions

The present work investigated for 3 months the presence of MPs in the influent and effluent of the two WWTPs in the city of Cadiz, including the evaluation of microplastics in the receiving water. The average abundance of MPs varied significantly in the WWTPs studied, along with the type of water received in the facility; in the case of the UWWTP the abundance of MPs was 645.03 ± 182.24 MPs/L and 16.40 ± 7.85 MPs/L in the influent and effluent, respectively. Whereas in the IWWTP, MPs concentration established was 1567.49 ± 413.18 MPs/L in the influent and 131.35 ± 95.36 in the effluent. These results evidence that IWWTPs present higher concentration of MPs than UWWTPs. Mean removal efficiencies at both WWTPs studied were higher than 90%.
Despite the high capacity to remove MPs shown by WWTPs, the relatively low concentration of MPs in the effluents of WWTPs combine with large sewage flow ($1.91 \cdot 10^7$ m3/year and $3 \cdot 10^4$ m3/year, in the UWWTP and IWWTP, respectively) arise to discharge considerable bulk of MPs into the receiving water. Estimating that UWWTP

can release up to 1.49 – 1.94 · 10^9 MPs/ day, whereas IWWTP drops approximately 1.07 – 2.64 · 10^7 MPs/day.

Regarding to morphological characterization, the most abundant length fraction was between 355 – 100 µm (> 50% in all the samples) and fibers were the amplest shape found in the present study, whilst chemical analysis the main types of MPs isolated from WWTPs were PVC, PE, HPDE in the urban plant and PVC, PA y EEA in the industrial plant.

The evaluation of the receiving water settled that MPs were more abundant in the Zone 2 (0.83 ±0.26 MPs/L) within the discharge point of industrial WWTP, than in Zone 1 (6.64 ± 2.71 MPs/L). Fibers were the predominant shape in the Zone 1, whereas in the Zone 2 fragments (possibly influenced for the industrial activity adjacent).

Respect to polymers identification, CA were the most abundant in the zone 1, whilst in the zone 2, PE and PP corresponded to the most abundant polymers.

To sum up, the present paper allows a deep knowledge of the occurrence, typology and removal efficiency of MPs in the wastewater treatment plants in the city of Cadiz and give an estimation of the amount of MP discharged into the environment by WWTPs, Finally, preliminary evaluation of these pollutants in the receiving water bodies was carried out, providing data to compare MPs presence in WWTPs and in the receiving water bodies.

Acknowledgments

This work was supported by the Spanish Ministry of Science, specifically via the project RTI2018-096771-B-I00, entitled ‘‘*Monitorización y análisis de toxicidad de microplásticos en EDARs. Aplicación de Tecnologías avanzadas para su eliminación*”. The authors wish to express their gratitude to Isabel Alarcón Carrasco.

Conflict of interest: the authors declare that they do not have any conflict of interest.

References

Alimi, O. S., Farner Budarz, J., Hernandez, L. M., & Tufenkji, N. 2018. Microplastics and Nanoplastics in Aquatic Environments: Aggregation, Deposition, and Enhanced Contaminant Transport. *Environmental Science and Technology, 52(4), 1704–1724. https://doi.org/10.1021/acs.est.7b05559*

Anderson, J. C., Park, B. J., & Palace, V. P. 2016. Microplastics in aquatic environments: Implications for Canadian ecosystems. *Environmental Pollution*, *218*, 269–280. https://doi.org/10.1016/j.envpol.2016.06.074

Asensio-montesinos, F., Oliva, M., González-leal, J. M., Carrizo, D., & Anfuso, G. 2020. Characterization of plastic beach litter by Raman spectroscopy in South-western Spain**.** *Science of the Total Environment*, *744*, 140890. https://doi.org/10.1016/j.scitotenv.2020.140890

Bretas Alvim, C., Mendoza-Roca, J.A. and Bes-Piá, A. 2020. Wastewater treatment plant as microplastics release source – Quantification and identification techniques. *Journal of Environmental Management, 255. 109739.*

Browne, M. A., Crump, P., Niven, S. J., Louise, E., Tonkin, A., Galloway, T., & Thompson, R. C. 2011. Accumulation of microplastic on shorelines woldwide : sources and sinks. *Environ. Sci. Technol. 45(21), 9175–9179* https://doi.org/10.1021/es201811s

Conley, K., Clum, A., Deepe, J., Lane, H., & Beckingham, B. 2019. Wastewater treatment plants as a source of microplastics to an urban estuary: Removal efficiencies and loading per capita over one year. *Water Research X*, *3*, 100030. https://doi.org/10.1016/j.wroa.2019.100030

Cristaldi, Antonio; Fiore, Maria; Zuccarello, Pietro; Oliveri Conti, Gea; Grasso, Alfina; Nicolosi, Ilenia; Copat, Chiara; Ferrante, Margherita. 2020. Efficiency of Wastewater Treatment Plants (WWTPs) for Microplastic Removal: A Systematic Review. *Int. J. Environ. Res. Public Health 17, no. 21: 8014.*

de Sá, L. C., Oliveira, M., Ribeiro, F., Rocha, T. L., & Futter, M. N. 2018. Studies of the effects of microplastics on aquatic organisms: What do we know and where should we focus our efforts in the future? *Science of the Total Environment*, *645*, 1029–1039. https://doi.org/10.1016/j.scitotenv.2018.07.207

Edo, C., González-pleiter, M., Leganés, F., & Fernández-piñas, F. 2019. Fate of microplastics in wastewater treatment plants and their environmental dispersion with effluent and sludge. *Environmental Pollution 259, 11383 https://doi.org/10.1016/j.envpol.2019.113837*

Elkhatib, D., & Oyanedel-craver, V. 2020. A Critical Review of Extraction and Identi fi cation Methods of Microplastics in Wastewater and Drinking Water. E*nvironmental Science & Technology.* 54, 12, 7037–7049. https://doi.org/10.1021/acs.est.9b06672

Franco, A. A., Arellano, J. M., Albendín, G., Rodríguez-barroso, R., Zahedi, S., Quiroga, J. Mª., Coello, Mª D. 2020. Mapping microplastics in Cadiz (Spain): Occurrence of microplastics in municipal and industrial wastewaters. *Journal of Water Process Engineering*, *38*, 101596. https://doi.org/10.1016/j.jwpe.2020.101596

Frias, J. P. G. L., Gago, J., Otero, V., & Sobral, P. 2020. Microplastics in coastal sediments from Southern Portuguese shelf waters. *Marine Environmental Research*, *114*, 24–30. https://doi.org/10.1016/j.marenvres.2015.12.006

Gago, J., Galgani, F., Maes, T., & Thompson, R. C. 2016. Microplastics in Seawater:

Recommendations from the Marine Strategy Framework Directive Implementation Process. *Frontiers in Marine Science*, *3*. https://doi.org/10.3389/fmars.2016.00219

Gatidou, G., Arvaniti, O. S., & Stasinakis, A. S. 2019. Review on the occurrence and fate of microplastics in Sewage Treatment Plants. *Journal of Hazardous Materials*, *367*, *504–512. https://doi.org/10.1016/j.jhazmat.2018.12.081.*

Gies, E. A., Lenoble, J. L., Noël, M., Etemadifar, A., Bishay, F., Hall, E. R., & Ross, P. S. 2018. Retention of microplastics in a major secondary wastewater treatment plant in Vancouver, Canada. *Marine Pollution Bulletin, 133, 553–561. https://doi.org/10.1016/j.marpolbul.2018.06.006*

Habib, R. Z., Thiemann, T., & Kendi, R. Al. 2020. Microplastics and Wastewater Treatment Plants — A Review. *Journal of Water Resource and Protection, 12, 1–35. https://doi.org/10.4236/jwarp.2020.121001*

Hann, S., Sherrington, C., Jamieson, O., Hickman, M. & Bapasola, A. 2018. Investigating options for reducing releases in the aquatic environment of microplastics emitted by products. *Final Report. Eunomia.*

Hidayaturrahman, H., & Lee, T. 2019. A study on characteristics of microplastic in wastewater of South Korea : Identification , quantification , and fate of microplastics during treatment process. *Marine Pollution Bulletin*, *146*, *696–702. https://doi.org/10.1016/j.marpolbul.2019.06.071*

Iñiguez, M. E., Conesa, J. A., & Fullana, A. 2017. Microplastics in Spanish Table Salt. *Scientific Reports, 7:8620.* https://doi.org/10.1038/s41598-017-09128-x

Iyare, P. U., Ouki, S. K., & Bond, T. 2020. Microplastics removal in wastewater treatment plants: a critical review. *Environmental Science Water Research & Technology 6, 2664-2675.* https://doi.org/10.1039/d0ew00397b

Lares, M. Chaker Ncibi, M., & Sillanpää , M. 2109. Intercomparison study on commonly used methods to determine microplastics in wastewater and sludge samples. *Environmental Science and Pollution Research 26:12109–12122.* https://doi.org/10.1007/s11356-019-04584-6

Li, J., Yang, D., Li, L., Jabeen, K., & Shi, H. 2015. Microplastics in commercial bivalves from China. *Environmental Pollution*, *207*, 190–195. https://doi.org/10.1016/j.envpol.2015.09.018

Liu, X., Yuan, W., Di, M., Li, Z., & Wang, J. 2019. Transfer and fate of microplastics during the conventional activated sludge process in one wastewater treatment plant of China. *Chemical Engineering Journal*, *362*, 176–182. https://doi.org/10.1016/j.cej.2019.01.033

Long, Z., Pan, Z., Wang, W., Ren, J., Yu, X., Lin, L., Lin, H., Chen, H., & Jin, X. 2019. Microplastic abundance, characteristics, and removal in wastewater treatment plants in a coastal city of China. *Water Research*, *155*, 255–265. https://doi.org/10.1016/j.watres.2019.02.028

Magni, S., Binelli, A., Pittura, L., Avio, C. G., Della Torre, C., Parenti, C. C., Gorbi, S., & Regoli, F. 2019. The fate of microplastics in an Italian Wastewater Treatment Plant. *Science of the Total Environment*, *652 (20)*, 602–610. https://doi.org/10.1016/j.scitotenv.2018.10.269

Masura, J., Baker, J., Foster, G., & Arthur, C. 2015. Laboratory methods for the analysis of microplastics in the marine environment: recommendations for quantifying synthetic particles in waters and sediments. *NOAA Technical Memorandum NOS-OR&R-48.*

Murphy, F., Ewins, C., Carbonnier, F., & Quinn, B. 2016. Wastewater Treatment Works (WwTW) as a Source of Microplastics in the Aquatic Environment. *Environmental Science and Technology*, *50* (11), 5800–5808.

https://doi.org/10.1021/acs.est.5b05416

Napper, I. E., Bakir, A., Rowland, S. J., & Thompson, R. C. 2015. Characterization , quantity and sorptive properties of microplastics extracted from cosmetics. *Marine Pollution Bulletin*, *99*(1–2), 178–185. https://doi.org/10.1016/j.marpolbul.2015.07.029

Napper, I. E., & Thompson, R. C. 2016. Release of synthetic microplastic plastic fibers from domestic washing machines: Effects of fabric type and washing conditions. *Marine Pollution Bulletin*, *112*(1), 39–45. https://doi.org/https://doi.org/10.1016/j.marpolbul.2016.09.025

Nel, H. A., & Froneman, P. W. 2015. A quantitative analysis of microplastic pollution along the south-eastern coastline of South Africa. *Marine Pollution Bulletin*, *101*(1), 274–279. https://doi.org/10.1016/j.marpolbul.2015.09.043

Ng, K & Obbard, Jeff. 2016. Prevalence of microplastics in Singapore's coastal marine environment. *Marine pollution bulletin. 52 (7). 761-767.* 10.1016/j.marpolbul.2005.11.017.

Norén, F. 2007. Small plastic particles in Coastal Swedish waters. *KIMO Report. 1-11.*

Ou, H., & Zeng, E. Y. 2018. Chapter 10. Occurrence and fate of microplastics in wastewater treatment plants. In *Microplastic Contamination in Aquatic Environments: An Emerging Matter of Environmental Urgency*. Elsevier Inc. https://doi.org/10.1016/B978-0-12-813747-5.00010-2

Plastics Europe Deutschland & Messe Düsseldorf 2019. Plastics - the Facts 2019. An analysis of European plastics production, demand and waste data. https://www.plasticseurope.org/application/files/9715/7129/9584/FINAL_web_version_Plastics_the_facts2019_14102019.pdf

Salvador, F., Turra, A., & Baruque-ramos, J. 2017. Synthetic fi bers as microplastics in the marine environment : A review from textile perspective with a focus on domestic washings. *Science of the Total Environment*, *598*, 1116–1129. https://doi.org/10.1016/j.scitotenv.2017.04.172

Sol, D., Laca, A., & Díaz, M. 2020. Approaching the environmental problem of microplastics : Importance of WWTP treatments. *Science of the Total Environment*, *740*, 140016. https://doi.org/10.1016/j.scitotenv.2020.140016

Sun, J., Dai, X., Wang, Q., van Loosdrecht, M. C. M., & Ni, B. J. 2019. Microplastics in wastewater treatment plants: Detection, occurrence and removal. *Water Research*, *152*, 21–37. https://doi.org/10.1016/j.watres.2018.12.050

Sundt, P., Syversen, F., Skogesal, O., Erik, P., & Hildonen, H. 2016. *Primary micoplastic-pollution: Measures and reduction potentials in Norway* (Issue April).

Talvitie, J., Heinonen, M., Pääkkönen, J. P., Vahtera, E., Mikola, A., Setälä, O., & Vahala, R. 2015. Do wastewater treatment plants act as a potential point source of microplastics? Preliminary study in the coastal Gulf of Finland, Baltic Sea. *Water Science and Technology 72. 1495-1504.* https://doi.org/10.2166/wst.2015.360

Talvitie, J., Mikola, A., Setälä, O., Heinonen, M., & Koistinen, A. 2017. How well is microlitter purified from wastewater? – A detailed study on the stepwise removal of microlitter in a tertiary level wastewater treatment plant. *Water Research*, *109*, 164–172. https://doi.org/10.1016/j.watres.2016.11.046

Thompson, R. C., Olsen, Y., Mitchell, R. P., Davis, A., Rowland, S. J., John, A. W. G., Mcgonigle, D., & Russell, A. E. 2004. Lost at Sea : Where Is All the Plastic ? *Science 304.* https://doi.org/10.1126/science.1094559

Mucha, M. (2015). Microplastics in Waste Water Treatment Plants and Separation Techniques. Diploma Thesis. Master Water Management and Environmental Engineering

Vuola, A.; Ruiz, M. and Vianello , A. (2019) FanpLESStic-sea 2019. Review of existing policies and research related to microplastics. *FanpLESStic-sea-project, 138pp*, doi: http://dx.doi.org/10.25607/OBP-824

Wagner, M., Lambert, S., 2018. Microplastics Are Contaminants of Emerging Concern in Freshwater Environments: An Overview. *The Handbook of Environmental Chemistry 58, 1-25.* https://doi.org/10.1007/978-3-319-61615-5_1

Xu, X., Jian, Y., Xue, Y., Hou, Q., & Wang, L. P. 2019. Microplastics in the wastewater treatment plants (WWTPs): Occurrence and removal. *Chemosphere*, *235*, 1089–1096. https://doi.org/10.1016/j.chemosphere.2019.06.197

Zhang, K., Shi, H., Peng, J., Wang, Y., Xiong, X., Wu, C., & Lam, P. K. S. 2018. Microplastic pollution in China's inland water systems: A review of findings, methods, characteristics, effects, and management. *Science of the Total Environment*, *630*, 1641–1653. https://doi.org/10.1016/j.scitotenv.2018.02.300

Ziajahromi, S., Neale, P. A., Rintoul, L., & Leusch, F. D. L. 2017. Wastewater treatment plants as a pathway for microplastics: Development of a new approach to sample wastewater-based microplastics. *Water Research*. 112: 93-99. https://doi.org/10.1016/j.watres.2017.01.042