Reuse of Water Contaminated by Microplastics

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Water treatment generally does not specifically address the removal of microplastics (MPs). Nevertheless, treatment plants process water effectively, and the number of synthetic microparticles in effluents is usually very low. Still, discharge volumes from water-treatment plants are often elevated (reaching around 10^8 L/day), leading to the daily discharge of a substantial number of MPs and microfibers. Furthermore, MPs accumulate in the primary and secondary sludge, which in the end results in another environmental problem as they are currently used to amend soils, both for cultivation and forestry, leading to their dispersion. Something similar occurs with the treatment of water intended for human consumption, which has a much lower but still significant number of MPs.

Keywords: wastewater ; sewage sludge ; microplastics ; microfibers

1. Introduction

The global demand for water is expected to increase by nearly one-third by 2050. Investing in water recovery and reuse could save up to 90% in energy and 70% in water usage, according to the United Nations (UN) [1]. Several of the Sustainable Development Goals (SDGs) are related to effective wastewater treatment. In objective 6, “Clean water and sanitation”, one of its goals is to improve water quality by reducing pollution, eliminating discharge and minimizing the emission of chemical products and hazardous materials, increasing safe reuse worldwide. Likewise, objective 11, “Sustainable cities and communities”, advocates for reducing the negative environmental impact per capita of cities, including paying special attention to air quality and the management of various toxic effluents.

Typical water-purification systems consist of pretreatment units (generally screening and clarification) and primary treatment that continues in a biological treatment unit (which constitutes secondary treatment). The primary treatment of wastewater consists of coagulation or flotation (for the removal of fats) followed by sedimentation. Afterwards, the organic matter in suspension is eliminated through a secondary treatment in the activated sludge reactor [2]. Many treatment systems also have tertiary units (often consisting of sand filtration or membrane-based filtration), ensuring a high-quality final effluent, whether for reuse in agriculture or safe disposal. Finally, a disinfection process is carried out (with chlorine, ozone or ultraviolet light) to eliminate pathogenic pollutants before discharging the treated water into the effluents. A simplified schematic representation of the processes that are generally present in a wastewater-treatment system is provided in **Figure 1**.

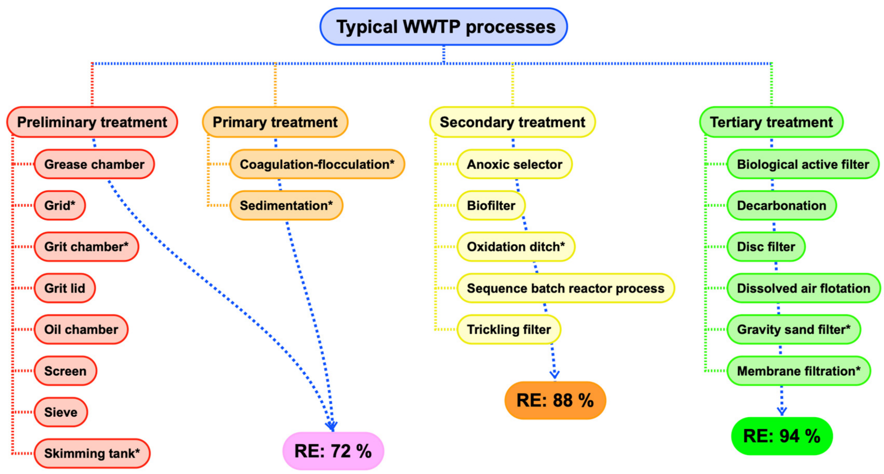

Figure 1. Main systems present in a wastewater-treatment plant (WWTP) [3]. Rough data on the removal efficiency (RE) of MPs of the different stages of treatment as a global, from Iyare et al. [4]. Those processes marked with an asterisk are present in the majority of plants.

If the treatment is focused on the purification of riverwater or water from desalination plants for human consumption, water-quality standards are much more stringent due to the expected lower concentration of foreign substances in drinking water. As a result, the preparation of drinking water generally requires the use of advanced water-treatment processes, such as membrane filtration and advanced oxidation. In fact, water and wastewater-treatment processes are similar, except for the biological processes that are required for wastewater due to the high content of organic matter [5].

Wastewater-treatment systems produce sludge, which must be treated before disposal or reuse [6][7]. The sludge needs to be concentrated to avoid the transport of extremely wet materials, among other reasons, and must also go through a process in order to destroy pathogens, and collect by-products such as biogas, in order to dispose of the sludge in an ecologically acceptable way [8].

On the other hand, as is well-known, plastics are present in almost all items related to human life, such as clothing, cosmetics, food packaging, equipment and instruments, etc. The annual global production of plastics has increased considerably in recent years, reaching 367 million tonnes in 2020 (only 1 million tonnes less than in 2019) [9]. It is important to add to these numbers the manufacture of synthetic fibers, since they are used in textiles, cords, fabrics, and other products, representing 61 million tonnes per year.

According to some estimates, around 10 million tonnes of plastic enter the oceans every year [10], accounting for 1.5 kg of plastic for each person on this planet, each year. Note that most plastics end up in sinks such as water collectors, rivers and seas until they drain into the oceans [11].

However, the problem of plastic is no longer that of “macroplastics” but of the plastic wastes being debased and reduced in size until they form what are called “microplastics”, i.e., tiny particles. Plastics, although resistant, are exposed to various factors that cause them to break into smaller pieces, producing microparticles that are present globally.

Microplastics (MPs) are generally understood to be plastic waste with sizes smaller than 5 mm [10]. The effect of human exposure to MPs is not yet known, which raises many unresolved questions [12]. For this reason, several studies are currently being carried out on the presence and influence of MPs, especially in the marine food chain, since when MPs reach the sea, they can be consumed by organisms such as zooplankton, thus entering the food chain.

Today, MPs are present everywhere, and come mainly from single-use plastics, fishing gear, clothing and cosmetics, paints, tires and urban dust [13]. These MPs easily reach rivers and seas due to their generally low density.

So far, three possible toxic effects of MPs have been indicated: the plastic particles themselves, their ability to adsorb persistent organic pollutants (POPs) and other substances present in aquatic systems around the world and the additives that these materials contain. Namely, the impact on human health stems from the fact that they can carry potentially toxic chemicals and microorganisms [14][15][16][17]. MPs could adsorb (or absorb) these dangerous chemicals, such as polyaromatic hydrocarbons (PAHs), metals and dioxins, from the environment and transport them to food products, which are ultimately consumed by humans [3][4][9][17].

MPs are classified as primary and secondary, as well as according to the polymer that constitutes them: polyethylene (PE), polyvinyl chloride (PVC), etc. Primary MPs are those that are already manufactured as MPs. More restrictions now apply to their manufacturing, but they have generally been used in hygiene products (exfoliating creams, toothpaste, gels, … usually known as personal care and cosmetic products, or PCCPs). Secondary MPs are those derived from the wear or degradation of large plastics (heat, UV, mechanical or biological degradation) [13][18]. Similarly, nanoplastics, with a size between 1 and 100 nm (i.e., 0.001–0.1 µm), can be produced by degradation of MPs or can be derived directly from industrial or domestic activity.

MPs are also classified according to their shape, and fall into the following categories (see **Figure 2**): fragments or microflakes (larger pieces, they could almost be called macroplastics); microfibers (with an elongated shape when observed under a microscope); microspheres or microbeads (which are small hard spheres); foams (sponge-like mass) and nurdles or granules (usually used in manufacturing). The terms nurdles and granules are used in many types of MPs when they do not fit in any of the others. The differences between pellets, nurdles, microbeads and microspheres are almost imperceptible. In much research, only the distinction between elongated microfibers and microparticles of a spherical or undefined shape is considered.

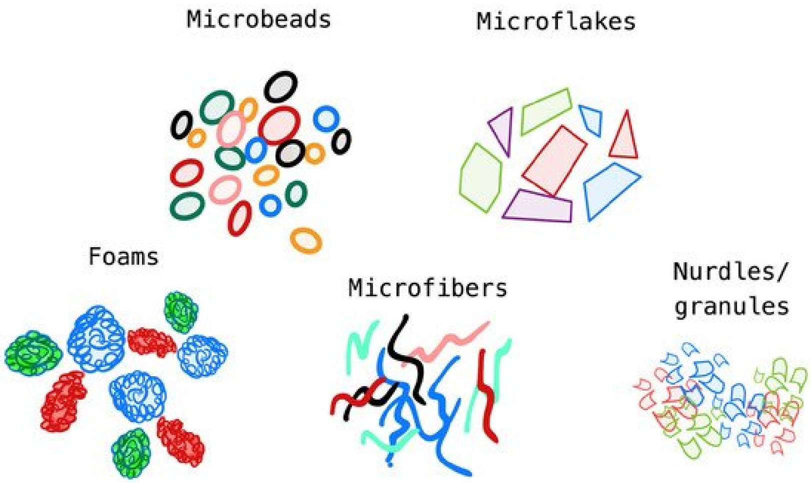

Figure 2. Main shapes presented by microplastics.

Each type of microplastic is associated with a main source. For example, microspheres are related mainly to the production of PCCPs, and microfibers are related to actions on synthetic polymers in clothing. These are considered the most common examples of microplastic contamination in the environment [14]. Fragments and fibers are the two dominant shapes of plastic particles in wastewater [15].

Nowadays, one of the sources of MPs entering the ocean are treated wastewaters. According to Bayo et al. [19] the effluent of a treatment plant can vary between 5900 MPs/m^{-3} on wet-weather days to 3000 MPs/m^{-3} on dry ones. As commented above, many synthetic-fiber fabrics (e.g., nylon, polyesters, polyacrylamides, etc.) derive from washing processes [15]. In this way, textile fibers, which account for around 70% of the MPs in WWTPs, are also commonly found in wastewater. Many clothes are made of polyester, which is a plastic very similar to polyethylene terephthalate (PET) (that of the majority of water bottles, for example) and it is already in the form of fibers, demonstrating the huge potential problem they can pose [10][16].

Murphy et al. [20] pointed out the problem represented by WWTP, which can be considered as sources of MPs in the aquatic environment. The study, performed in a plant in Glasgow, showed that the removal efficiency of MPs was higher than 98%, but still the release of 65 million MPs into the receiving water was denounced.

The main findings related to the elimination of MPs in wastewaters is carried out, with special interest in elimination by filtration. The removal efficiency of several systems will be discussed, as well as the effect of the shape and size of the MPs to be eliminated and the nature of the polymers.

2. Microplastic Fate in Conventional Water/Wastewater-Treatment Processes

The fate of MPs entering WWTPs is currently a very relevant research topic. Many authors have investigated the role of MPs in the different stages of wastewater treatment. It has been observed that the presence of MPs reduces the efficiency of wastewater and sludge-treatment processes and increases sludge volume [6][21], affecting methane production in bioreactors, for instance. The presence of MPs could make it necessary to change the configuration of the treatment plant. For example, the presence of a large number of MPs could oblige to the use of certain reagents. Similarly, a decrease in the nitrogen conversion rate and fouling of the membrane in wastewater-treatment processes have been described [22]. As a result, it is difficult to accurately assess MP-removal efficiency (RE) in WWTPs. At present, there are no policies or legislation that require a minimum for MP removal during wastewater treatment.

Johnson et al. [23] looked for the presence of MPs in potable water. They found extremely low amounts, less than 2 particles/m^3, showing that there was an effective removal of this type of particles in the water-treatment plants.

In their review, Liu et al. [24] presented the numbers of MPs in the influent of different WWTPs. They showed that the abundance of MPs in the influent was between 0.28 and 31,400 MPs/L, while that in the effluent was between 0.01 and 297 MPs/L. They found quantities of MPs in the sludge varying within the range $4.40{\cdot}10^3$–$2.40{\cdot}10^5$ MPs/kg.

Rasmussen et al. [25] calculated the masses of plastics of >500 μm in a WWTP. They showed that the plant load was 202.2 kg/day, of which 73% was retained. The remaining plastic was found in the sludge (13.6%) and in the effluent (0.4%).

Tadsuwan et al. [26] pointed out that the RE of a grit trap alone was >33%, followed by 14.2% removal in a secondary treatment. In the sludge, the number of MPs was 8120 particles/kg.

Even considering that conventional WWTPs usually eliminate more than 90% of the incoming MPs, these systems continue to be the main source of introduction of MPs into the environment, as the high volumes of generated effluents represent a very large contribution [27]. Simon et al. [28] noted that Danish WWTPs discharge around 3 t/year of MPs in the size range of 10–500 µm, despite the 98% RE of this size of particles. Bretas-Alvim et al. [27] presented data on the discharge of MPs in different countries, observing important differences (±1000% approx.) among them.

Recently [29] a study investigating the MP removal at different treatment stages showed that the primary treatment process has an effectiveness of about 75%, and that it increases to 91.9% in the secondary stage. Removal efficiency further increased to >98% after tertiary treatment (coagulation, ozonation, disc membrane filters and rapid sand filters).

Iyare et al. [4] studied the fate of MPs in 21 WWTPs around the world. The authors show that the preliminary and primary treatment of wastewater remove 72% of the MPs, and that the removal efficiency increases to 88% (of the number of microplastic particles present in sewage influent) in secondary treatment and 94% in the tertiary. A major part of the MPs eliminated are accumulated in the sludge, which has lately been used for soil enrichment, causing accumulation in terrestrial ecosystems [4]. Ou and Zeng [30] pointed out the minor presence of MPs at night, showing an uneven temporal distribution of MPs.

The largest WWTP in P. R. China was studied by Yang et al. [31]. A concentration of approx. 12 MPs/L was detected in the influent, reducing the amount to 0.6 MPs/L by employing a conventional WWTP that was equipped with ultrafiltration as the final tertiary treatment. This study shows a decrease in the MP concentration throughout the treatment processes, i.e., the RE in the primary treatment was ca. 59%; it was 72% after the secondary stage; and that of the overall process was 95%. This research also investigates the role of the MP size and found an entering average size of ca. 1110 µm, being quite similar to the size in the effluent. The main polymers detected, as in many other studies, were PET, PS (polystyrene) and PP (polypropylene). The fact that they find similar sizes at the inlet and the outlet of the plant after an ultrafiltration process suggests that MPs might not be properly retained in the filter, or that a mix of filtered and unfiltered waters was produced.

Talvitie et al. [32] investigated the behavior of advanced treatment technologies in various WWTPs. The highest removal efficiency was achieved by the membrane bioreactor (MBR) with a RE of 99.9%, followed by gravity or rapid sand filter (RSF) (RE = 97%) and air flotation (RE = 95%).

In the study led by Lv et al. [33] two parallel systems of wastewater treatment were studied, namely oxidation ditch treatment and MBR. The MPs observed by these authors at the plant entrance consisted of PET (47%), PS (20%), PE (18%) and PP (15%). Regarding the shape of the microparticles, a greater proportion of fragments was found (65%), followed by fibers (21%, mainly PET), films (12%) and foams (2%). No microbeads were observed. Regarding the size of the particles, the dominant size was >500 µm (40%) and between 62.5–125 µm (29%). These researchers observed that MP concentrations increased in the treatment systems as a function of the ease of the treatment process. In this way, the MPs were eliminated by 99.5% in the MBR system compared to 97% in the oxidation system (by weight). These percentages indicate the tendency to retain the largest fragments.

The study developed in Finland by Talvitie et al. [34] also showed the effectiveness of WWTPs for the elimination of MPs. The results show that conventional primary and secondary treatments can effectively eliminate (>99%) the MPs that reach the WWTP. The majority (98%) of the MPs were eliminated during the primary treatment. The activated sludge (secondary) process further decreased (~88% from process input) the MP concentration. During wastewater treatment, most MPs (>99.5%) were retained in the primary and secondary sludge; however, part (~20%) of the retained MPs was recirculated back to the treatment process as part of the water was rejected.

The authors [34] stated that the removal of MPs can be further improved with the implementation of a specific treatment system. The proposed equipment consists of a membrane bioreactor (additional RE = 99.9%), sand filtration (RE = 97%), dissolved air flotation (RE = 95%), or disc filtration (RE = 40–98.5%). Biologically active filtration had no impact on the concentration of MPs. Nevertheless, Bayo et al. [12] found persistent low-density polyethylene (LDPE), nylon and PVC in the RSF and melamine after MBR treatment.

Gies et al. [17] studied the presence and removal of MPs in a Canadian WWTP. They found that only 32.4% of the suspected MPs were plastic polymers, and that the global removal efficiency of the plant varied between 97.1% and

99.1%. Nevertheless, the number of MPs released to the environment was concentrated in the primary and secondary sludges.

Ben-David et al. [35] studied the elimination of MPs in a WWTP sited in northern Israel, observing a removal efficiency higher than 97%, with most MPs eliminated in the secondary stage and observing an average of 1.97 MPs/L after sand filtration.

Carr et al. [36] showed that existing treatment processes are effective at removing MPs in WWTPs, and found no MP presence after tertiary treatment in seven plants, and only 0.00114 MPs/L for effluent after secondary treatment. In this way, MP particles were found to be removed mainly in the primary treatment zones during the processes of solid skimming and sludge settling. These authors mention that some consumer products, as the toothpaste containing MPs may be contributing much more than others to the amounts of MPs reaching the WWTPs.

Cheng et al. [3] found a global removal efficiency of MPs higher than 93%, by weight, going from 61–5600 µg/L to 0.5–170 µg/L. These researchers showed that preliminary and primary treatments contribute to the removal of MPs, and that the efficacy of secondary and tertiary treatments was, understandably, highly dependent on the applied techniques.

Cristaldi et al. [15] reviewed information on the removal efficiency of different WWTPs around the world, showing a removal efficiency greater than 90%. This seems to be a good situation, but the high amount of water treated in the plants means that millions of MPs continue to be released every day into the aquatic environment.

Biofilters have also been tested for the removal of MPs from treated wastewater. In their study, Liu et al. [37] showed that a raw effluent containing 0.917 MPs/L was easily filtered to 0.179 MPs/L in the first stage of a conventional biofilter. Nevertheless, small sizes, as expected, cannot be completely removed, although no MPs higher than 100 µm in size were found (i.e., RE = 100% for sizes lower than 0.1 mm).

Ngo et al. [38] showed that the existing WWTPs are inefficient to completely remove the MPs and there is a risk that they may be discharged into the ambient water.

In many studies, filtration has been shown to play an important role in removing MPs. However, this method has its drawbacks [21][39] because the mechanical stress generated during treatment can cause plastics to wear away, resulting in smaller particles that are likely to be released into the environment without restriction.

2.1. Microplastic Accumulation in Sludges

Zhang et al. [6] insisted that in a WWTP, microplastics are simply transferred from sewage to sludge; this could be a problem during anaerobic digestion, as MPs can contain significant amounts of toxic substances such as persistent organic pollutants. Gatidou et al. [40] showed that MP content ranges up to 3160 MPs/L in raw water, 125 MPs/L in treated wastewater and $170.9 \cdot 10^3$ MPs/kg total dry solid in the sludge. Gies et al. [17] observed that primary sludge had approx. $14.9 \cdot 10^3$ MPs/kg, with fibers being more abundant than other particles. Likewise, secondary sludge had $4.4 \cdot 10^3$ MPs/kg, with fibers also being the most abundant shape. The dominance of fibers in sludge samples has been previously reported [41][42]. On their hand, Liu et al. [24] indicated that the microplastic abundance in the sludge was within the range of $4.40 \cdot 10^3$–$2.40 \cdot 10^5$ particles/kg.

Because MPs accumulate in biological sludge, and because these sludges are mainly used to improve agricultural or forest soil, studies should be carried out on the incorporation of these particles in meat from land animals [27].

Regarding the MP content in sewage sludges (SS), Ou and Zeng [30] indicated that the grease on the SS samples contained a number of MPs that was significantly higher than the grit and sludge-cake samples. PE microbeads from PCCPs were dominant in grease samples.

Habib et al. [43] observed an abundance of synthetic fibers in soil conditioners, fertilizers and similar biosolid materials, denouncing the presence of MPs in sludges.

Jiang et al. [44] studied the fate of MPs in sewage and sludge filter cake. These authors showed that there were 126.0 MP particles/L in the influent and 30.6 in the effluent, transferring about 75% of the MPs to the sludge. In their study, the abundance of MPs in dewatered sludge and filter cake was 36.3 and 46.3 particles/g. These figures are equivalent to the accumulation of $7.74 \cdot 10^{12}$ particles/year in the sludges.

This implies that most of the MPs removed during wastewater treatment will end up in the open environment [35][45]. For this reason, Wolff et al. [46] indicated that WWTPs are a good sink for MPs only when the sludge is thermally treated and not used for agricultural amendment. Incineration and production of chemicals by pyrolysis of sludges from WWTPs are very promising current lines of research [47] that would avoid the accumulation of MPs in the environment. These techniques would, furthermore, reduce the energetic demand on the treatment plants, increasing the potential for chemical-energy recovery from wastewater pollutants [48].

In this way, an alternative route for disposal, incineration of SS with heat recovery, has been pointed out as a good practice [49]. Note that in the European legislation [50] incineration is preferred to landfilling. This is because landfill is much less controllable, producing gas release and in many cases spontaneous combustion of the biogas, in addition to the fact that the necessary technology for the treatment of the gas produced in the incinerator is already available. Through incineration, MPs contained in the SS would definitively be eliminated from the environment, in what emerges as a safe alternative for disposal.

2.2. Need for Standardized Methods

There is no standard protocol for MP measurement in WWTPs. Many authors [3][27][51][52][53][54] insist that the differences between studies are related to the different analytical techniques employed for measuring MP contamination.

Lusher et al. [55] insisted on the necessity of harmonization in the isolation and extraction of MPs from environmental samples, including the sample-collection techniques and data analysis. On the other hand, Oβmann [56] insisted that the harmonization of methods for the quantification of MPs in drinking water and other food should be developed and defined.

Bayo et al. [19] indicated that the concentrations reported by different authors on the presence of MPs in WWTPs are difficult to compare, because different sampling techniques, volumes and analytical methods are used. Furthermore, the use of MP concentrations on a numerical basis has been criticized for overstating MP abundance because this involves counting the number of suspected particles [3].

Sun [57] pointed out that since MP sampling and detection methods can significantly affect the result of their quantification and identification, harmonization is urgent. Hong et al. [58] proposed a method for estimating the mass of MPs in sewage, based on the quantification of total organic carbon (TOC).

Moreover, differences can be found due to the difficulties present in the sampling of MPs in waters. Lenz et al. [59] designed a water-sampling system specially dedicated to the collection of MPs in surface and groundwater for environmental pollution studies. It is designed specifically for sampling small MPs (size fraction <300 μm) in different source areas. Li et al. [60] mentioned that in many MP studies, these particles are just sampled by nets with a typical mesh size of 330 μm. The sampled MPs are usually purified and digested, and lately analyzed by different methods, but there are no universally accepted sampling and quantification tools, so important differences among studies can be found.

Another interesting topic is the treatment of samples in the laboratory. Elkhatib et al. [53] insisted on the need for quality-assurance methods. This would include the use of a wet filter exposed to air during sample processing to take into account air contamination, the analysis of blank samples using deionized water, the addition of a sample with a known polymer, and the analysis of samples in triplicate. Ou and Zeng [30] showed that the use of H2O2 to preoxidize the particles can remove surface biogenic materials from the polymer. However, the preoxidation combined with heating induced the melting of some MPs, especially microbeads. Dyachenko et al. [61] also used peroxide oxidation to eliminate interferences in the analysis.

Gatidou et al. [40] also insisted that special care should be taken on the types of clothing used. Gies et al. [17] observed a deposition of 36 fibers on a 1 μm pore-size × 47 mm-diameter filter membrane over an 8 h period.

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