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The occurrence of microplastics in wastewater and the possibilities of using separation methods to reduce this contamination at the wastewater treatment plant

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ABSTRACT

Microplastics are small pieces of plastic that accumulate in all elements of the. They are created as a result of fragmentation of larger pieces of plastic or they are introduced into the environment as primary microplastics. Due to their small size, microplastics are considered to be bioavailable for organisms at all trophic levels. Their structure makes them susceptible to the adhesion of organic pollutants and the leaching of substances considered to be toxic. In this way, microplastics became a serious threat to the aquatic environment, and thus to the organisms living in it and to the further trophic levels. At present, it is little known to what extent treated and discharged sewage affects the accumulation of microplastics in the environment and how these pollutants behave during transport through wastewater treatment plant (WWTP) facilities. The study showed that the treatment plant contributed to the removal of microgranules. However, the amount of microfoils and microfibers increased in the subsequent stages of the technological process. Although WWTPs use multi-stage wastewater treatment technologies, they are not properly designed to remove microplastics, and WWTPs are reported as an important source of microplastics in water. One of the solutions to change this is the use of specific solutions represented, among others, by separation methods, such as tertiary wastewater treatment in integrated wastewater treatment systems. Therefore, tests were carried out to identify microplastics in wastewater at various locations in the wastewater treatment process at the WWTP. Then, the test results were used to select separation methods that would allow reducing the number of microplastics at individual stages of the technological process, in particular at the outflow from the WWTP. The currently used methods of municipal wastewater treatment are insufficient. Therefore, one of the aims of this publication is to present the possibilities of using other, additional and more effective technologies.

Keywords: Microplastics removal; Granular particles; Fibers; Wastewater treatment plant; Wastewater treatment; Separation processes

1. Introduction

In 2019, the European Plastics Industry employed over 1,500,000 people in over 55,000 companies and generated around EUR 350 billion of turnover, and global plastic production reached almost 368 million tons, increasing by 2.45% over the previous year. European production amounted to 57.9 million tons, which is about 6.3% lower than in 2018, but the world production is still increasing (in 2019 – 368 million tonnes while in 2018 – 359 million tonnes), especially due to production in Asia (more than half of the world production) [1]. Such a huge production

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is not fully utilized and the percentage of plastic recycled is only 32.5% [1]. Unfortunately, most of the unutilized plastic goes to the environment where it is fragmented and becomes microplastic. The primary microplastics (e.g., produced for cosmetic purposes, present in cleaning agents, and toothpaste) also flow into the raw wastewater. Together, the primary and secondary microplastics could be dangerous and difficult to get rid of, even in technological processes at wastewater treatment plants (WWTPs).

Microplastic particles can enter the hydrosphere in many different ways. One of the most obvious forms of getting this pollution into water is wastewater outflow [2–4]. Microplastics in wastewater can come from the aforementioned cosmetic products, detergents, and also from rubbing synthetic fabrics against each other during washing. Plastics can also easily reach industrial wastewater, for example, in plants using the plastic shot for sandblasting, plants that use chemicals producing plastic microparticles, or produce such agents [5]. Although most of the microplastics in WWTP are removed, they remain an important source of this pollution. There are many examples in scientific literature. Nocoń et al. [2] found that in the Kłodnica River below the discharge from WWTP the concentration of microplastics is higher. In the effluents from Lower Saxony WWTP microplastic particles lower than 500 m were found even after post-filtration [6]. Murphy et al. [7] showed that despite high retention of MP in WWTPs, a considerable number of these particles enter the environment due to the high flux of wastewater. The shipbuilding industry turns out to be another important source. Here, plastic dust often results from cutting or grinding plastic parts. Microplastic particles can also be washed out of varnishes and paints covering ship hulls, buoys, and elements of hydrotechnical infrastructure [8].

These processes are influenced by the conditions the material is exposed to (e.g., whether the molecule in question is in an aquatic or terrestrial environment), the properties of a given polymer, including its density, crystallinity, type, and content of modifying additives [5]. These processes cause the plastic to lose its plasticity. In the natural environment (aquatic and soil), this usually occurs due to UV radiation or hydrolysis [9]. Such weakened fragments of the material are then crushed due to friction forces arising, for example, as a result of rubbing against stones or other obstacles while moving down the river or rubbing against each other. The particles can also become disintegrated by waves, the screws of passing ships, crushed by insects or torn apart by larger creatures [8]. Under these conditions, the complete degradation of plastic materials, for example, bottles will take a long time.

Plastic impurities in their primary form, or the form of microplastics, can end up in the aquatic environment not only as a result of direct emissions to water but also as a consequence of migration processes from the terrestrial environment. It is estimated that as much as 75%–90% of the microplastics in water come from land sources [8]. Agriculture makes a huge contribution to the penetration of microplastics. Plastic is used for the coverings of greenhouses or fertilizer sacks; huge amounts of plastic mulch are used for crops [8] and silage for animals is packed in foil. All these elements can break down into microplastics. As previously mentioned, a significant part of the microplastic material is removed in WWTP. According to researchers, it can even be 72%–98% [10]. WWTPs intensify the phenomena occurring in nature. Most anthropogenic pollutants in municipal wastewater are biodegradable. In the case of plastic, one can only observe the transformation of macroplastics into microplastics. A more important process is the physical separation of this pollutant. Sludge from the wastewater treatment plants, containing microplastics retained in the WWTP, is then used on the arable fields as a fertilizer. Waste landfills are another path for plastics to find their way into the environment. The problem is that landfills are not adequately protected against the effects of natural forces, such as wind or heavy rains. A large group of waste comes from building and construction products, such as pieces of foamed polystyrene or foil. The global road and transport system contributes to the problem through abrasive dust from vehicle tires and paints used for painting belts [5]. There are many different ways in which these impurities can end up in the water. Their characteristics such as low weight, durability, and shape of some particles allow them to be moved over great distances [11]. Particles lying on roads, arable fields, construction sites, or unsecured landfills can be moved by the wind and lifted into the upper atmosphere. Then, they can fall onto the water surface alone or together with precipitation [12]. Such precipitation is a very important source of microplastics in aquatic environments and should be taken into account in all studies on microplastics and waters [13]. The microplastic particles can be flushed from land to surface and underground water by water runoff during rainfall and snowmelt [14]. The flow of plastic particles with water is conditioned by the type of terrain surface, and certain tendencies, such as frequent mowing of grass (e.g., on roads), significantly facilitate it [5]. Once in the aquatic environment, plastics continue to migrate over long distances along with river and sea currents. Research indicates that transport in an aquatic environment is not limited to a horizontal plane. A significant part of the microplastics can sink deeply below the water surface, which is confirmed by the content of microplastic particles in oceanic samples taken from a depth of up to 5,000 m [15]. Migration processes can also be of anthropogenic origin. The plastics can be moved and crushed by the previously mentioned mowing of grass or washing the surface of roads and squares [5]. Very large amounts of microplastics, or impurities that will become microplastics in the future, may end up in the aquatic environment due to unpredictable phenomena, such as natural disasters in the form of tornadoes, floods, and even fires.

As the examples cited show, there are a very large number of ways for microplastics to get into the environment. The number of sources of this pollution is reflected in research results from around the world. In 2015, scientists estimated that 5 billion plastic particles are floating in the oceans, of which more than 90% were microplastics [15]. The concentration is variable and depends on the place on the ground where samples were taken. In the Arctic, it is several particles per kilogram of seawater [10], in the Eastern Atlantic in equatorial regions – 13.5 particles/kg, while in the Italian Coast as many as 2,175 particles/kg [9]. In the Polish coast (southern Baltic Sea) the mean concentration of microplastics varied between 76 and 295 particles/kg of dry sediment, with fibers and plastic fragments being the most dominant [16].

As for rivers, Rhine water samples showed an average microplastics content of 892,777 particles/km^2, with a maximum value of up to 3.9 million particles/km^2 [5]. In the Swiss Sections of the Rhone, Aubonne and Venoge concentrations ranged between 0.10 and 64 particles/m^3, in the Chinese mouths of the Jiaojiang, Oujiang and Minjiang rivers, located in urban areas, the content of microplastics was higher and amounted to 100–4,100 particles/m^3 [9]. Studies carried out on the water from the Swiss Lakes: Geneva, Bodensee, Neuchâtel, Maggiore, Zurich, and Brienz showed microplastics contents between 11,000 and 222,000 particles/km^2 [9]. In the Great North American Lakes on the United States–Canada border, the average content was 43,000 particles/km^2. However, samples taken near cities showed a content of up to 466,000 particles/km^2 [17]. Samples from Lake Taihu in China showed a content of 6,000,000 particles/km^2. By comparison, Lake Hovsgol, located in a sparsely populated area of Mongolia showed a content ranging from 997 to 4,435 particles/km^2 [18].

Due to the high amounts of microplastics shown in the above surface waters, it is necessary to trace the sources. Wastewater treatment plants play an important role in the microplastic contamination of surface waters [19,20]. During conventional wastewater treatment, mainly treated wastewater has a smaller microplastics load than the wastewater flowing into the treatment plant. This is due to the accumulation of microplastics in the sludge. However, according to some scientific reports, the concentration of synthetic microplastics in treated wastewater is higher than in wastewater before the treatment process. Researchers indicate that the problem may lie in the failure to adapt the used treatment technology for the removal of these specific impurities.

The aim of the paper is to determine the changes in the amount of microplastics in wastewater at various technological stages of municipal wastewater treatment. In the paper, the authors also analyze the possibility of using separation methods to reduce the amount of microplastics in treated wastewater.

2. Materials and methods

The tests were carried out in WWTP of a city located in the Silesian Agglomeration (Poland). The average daily load of WWTP A is approx. 18,000 m^3/d (PE equal to 77,000). There is a sanitary, rainwater, and combined sewerage system in the WWTP catchment area (about 66.1 km^2). WWTP is used primarily for urban wastewater, but it also treats industrial ones coming from industrial plants located in the catchment area.

The flow diagrams of the treatment plant with sampling points are presented in Fig. 1. WWTP is a mechanical-biological installation. The hydraulic retention of wastewater in biological reactors is about 74 h, and in the secondary sedimentation tank, it is about 12 h. It is worth noting that the tested WWTP does not have the primary sedimentation tank.

The research consisted of the quantification of microplastics at several different characteristic points on various treatment stages of WWTP. Samples were taken: below the grid chamber, in the secondary sedimentation tank, at the WWTP effluent.

Three series of simultaneous measurements were carried out in WWTP in the spring and summer of 2019. Wastewater samples with a volume of 50 L were taken at the above-mentioned sampling points. The random wastewater samples were then passed through the plankton net (0.25 mm mesh size). To clarify the sample and decompose organic matter, hydrogen peroxide (30%, 25 cm^3 per sample) and iron sulfate (1 g per sample) was added to the samples (iron acts as a catalyst here) and heated at a temperature not exceeding the boiling point of water. The heating process was conducted at 70°C. After heating to

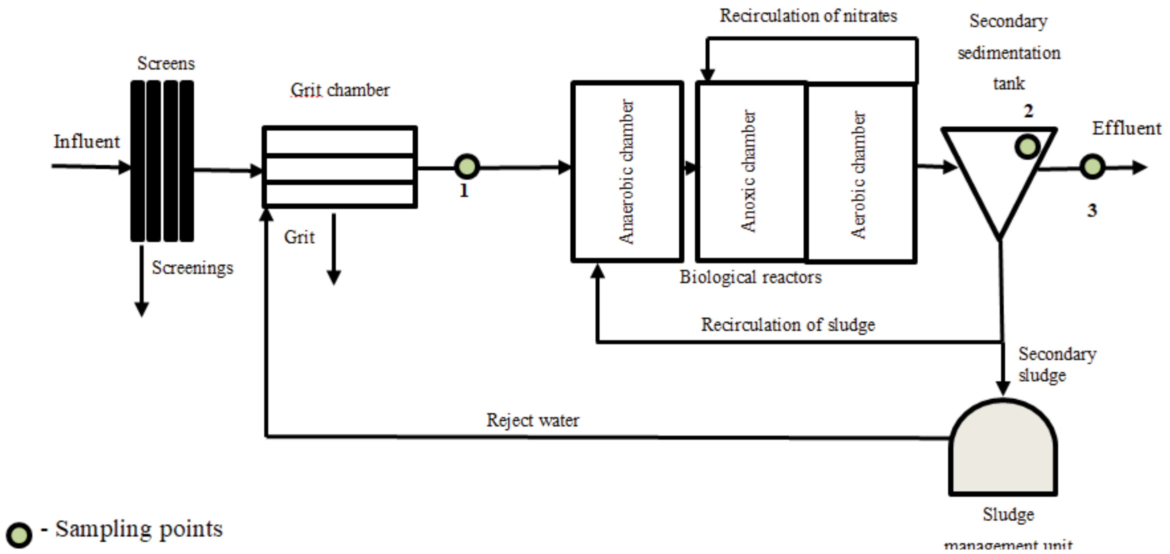

Fig. 1. Flow diagrams of wastewater treatment plant with sampling points.

identify microplastic particles, the samples were examined under a Delta Optical SZH – 650 B/T microscopes. Thanks to the use of a camera coupled with a microscope, the observations were carried out on a computer screen using the ScopeImage 9.0 program. The performed activities consisted of counting microplastic particles, also with the use of Thom's chamber.

3. Results

The tests were carried out for the quantitative determination of microplastics contained in wastewater and their grouping by form in which they occur. Table 1 summarizes the average concentrations of microplastics with SD and RSD values.

Statistical parameters show that the amount of microplastics flowing into the biological part is variable over time. Such variation can also be observed at other wastewater collection points. However, there is a clear increase in the number of microplastic particles between the secondary sedimentation tanks and the effluent (samples were already taken on the border of the WWTP). These differences may also indicate significant diurnal changes in the amount of plastics flowing to the WWTP. According to the authors, the number of microplastics in the surface layer of treated wastewater in the secondary sedimentation tank may also be influenced by operating conditions. The WWTP operates in the area of mining activities, which is the reason for changes in the terrain surface. Uneven settlement of such large building structures is a typical phenomenon for the Upper Silesian Agglomeration, and within the secondary sedimentation, it is often the cause of uneven hydraulic loading of overflow edges and the formation of places where the water is stagnant.

In the samples taken from the grit chamber 1,600–3,880 particles/m^3 of microplastic elements were collected, with microfibers being the most dominant – the average amount was about 40% for each series. In the secondary sedimentation tank, with a total number of microplastics amounting to 3,040–4,000 particles/m^3, the largest number of microfibers was also found – the average amount was about 46% for each series. The number of microplastics has increased during the technological process, which may mean that plastic can be fragmented into smaller parts increasing the migration of microplastics at each subsequent stage of the wastewater treatment process. It was noted that the largest number of microplastics was found in samples taken at the outflow of the treatment plant, 2,460–5,920 particles/m^3. Here, microfibers were also the most dominant. Their average amount was about 50% of the total amount of microplastics. Greater amounts of microfoils and microfibers in the effluent than in the secondary sedimentation tank may indicate the decomposition processes of organic matter at the bottom of the sedimentation tank and, consequently, the release of gases as products of these processes. Bubbles of these gases can stick to microfoils and microfibers and carry them to the effluent. The largest amount of microplastics in the WWTP effluent may be associated with the insufficient treatment of wastewater that does not remove very small size pollution. Microplastics smaller than 20 μm and nanoplastics are not retained from WWTP [21]. The treated wastewater from a conventional WWTP in Denmark contained 917 microparticles/m^3 which corresponded to a mass concentration of 24.8 μg/m^3 [22], in the USA it was only 50 particles/m^3 [23]. In turn, plastic contaminants larger than 0.7 mm (micro- and nanoplastics), 9,000–91,000 particles/m^3 were found in an outflow in the Netherlands [23].

According to Lares et al. [24], there are two types of microplastics occurring in wastewater: microplastic particles and fibers. However, in this study, they were divided into two additional categories: microfoils (Fig. 2), microfibers (Fig. 3), microgranules (Fig. 4), and unidentified (Fig. 5). Unidentified objects are those that show the properties of plastics but do not fit into other categories.

Microgranules can be considered as primary microplastics, for example, produced for cosmetic purposes, present in cleaning agents, and toothpaste, most often made of polyethylene or polypropylene. The most common microplastics in wastewaters are fibers (Fig. 3).

Table 1
Microplastics in samples taken from wastewater treatment plant

Place of sampling		Foil particles (m^3)	Unidentified particles (m^3)	Fiber particles (m^3)	Granule particles (m^3)	Total particles (m^3)
Grit chamber	Range	320–680	40–720	720–1,480	520–1,000	1,600–3,880
	Medium	487	313	1,047	787	2,633
	SD	181	359	391	244	1,155
	RSD (%)	37	115	37	31	44
Secondary sedimentation tank	Range	880–1,020	320–520	1,200–2,040	560–680	3,040–4,000
	Medium	927	420	1,700	627	3,673
	SD	81	100	442	61	549
	RSD (%)	9	24	26	10	15
WWTP effluent	Range	820–2,260	240–480	1,320–2,800	80–480	2,460–5,920
	Medium	1,673	340	2,273	300	4,587
	SD	756	125	827	203	1,861
	RSD (%)	45	37	36	68	41

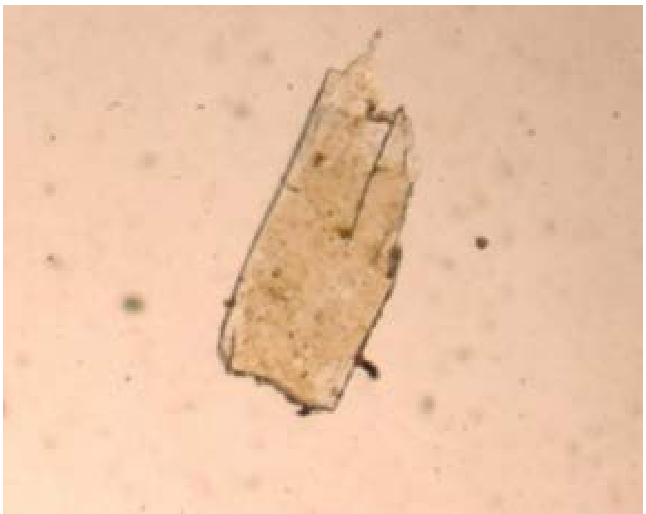

Fig. 2. Exemplary microfoil found in wastewater (magnification 40×).

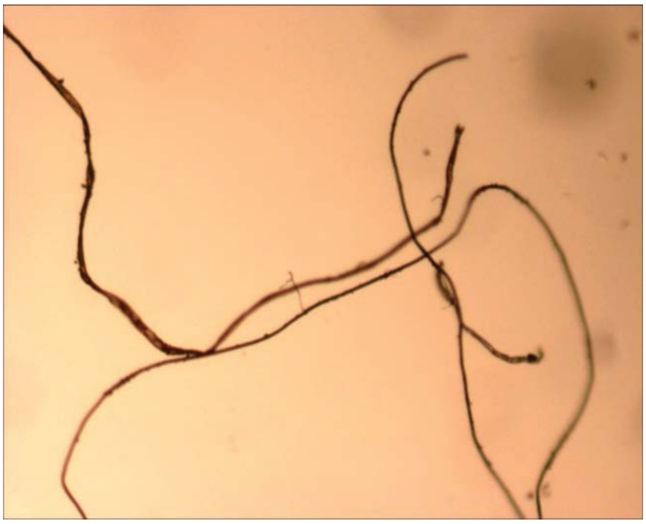

Fig. 3. Exemplary microfibers found in wastewater (magnification 40×).

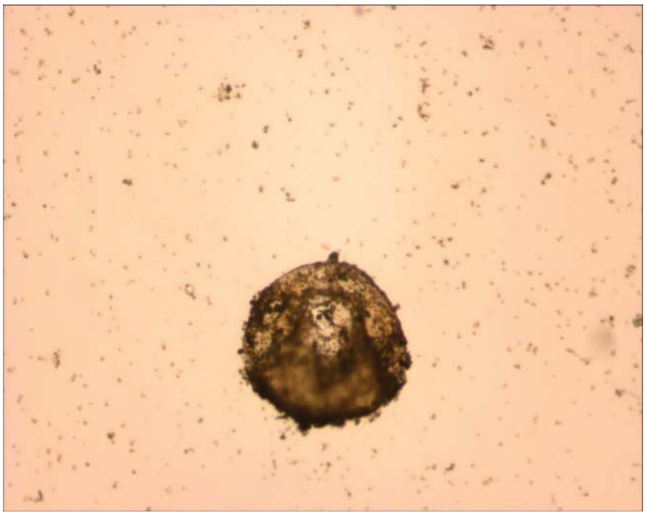

Fig. 4. Exemplary microgranule found in wastewater (magnification 40×).

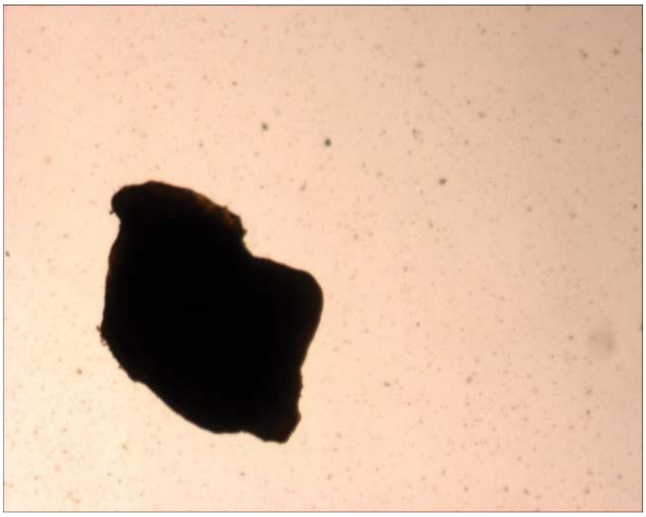

Fig. 5. Exemplary unidentified microplastic found in wastewater (magnification 40×).

The microfibers are most often made of polyester and can come, for example, from drains of washing machines [25]. However, it should be mentioned that there is a large amount of industrial, mining, and medical wastewater flowing in. All medications and dressings after fragmentation can be considered as microplastics, in particular microfibers. A large number of microfibers in this research may indicate a significant percentage of municipal or industrial medical sewage from a nearby large medical center and two hospitals, which are the source of textiles in the WWTP.

Many authors in their scientific research [5,8,9,12,14] indicate that municipal WWTPs are a significant source of microplastics in the aquatic environment and similar conclusions can be drawn from the results in Table 1. It should also be taken into account that virtually all WWTPs require at least one pump to be used. It is the pump impellers that contribute to the grinding of larger plastic fragments that flow into WWTP. Taking into consideration all of these wastes, research indicates that they are often eliminated in amounts significantly greater than 90%. It is also worth noting that none of the WWTPs operated in Poland were designed to remove microplastics. The need for this type of solution is confirmed by the results presented below. Based on the test results obtained in the study, the increasing quantity of microplastics in the wastewater treatment process was also estimated.

Table 2 summarizes the amounts of microplastic particles by type and percentage increase of microplastics quantity in the wastewater treatment process at WWTP. For a reliable comparison, the results include the samples taken after the grit chamber and from the effluent, that is, the first and last point from which sampling was possible.

Table 2
Increase of microplastics quantity during the wastewater treatment process in wastewater treatment plant

Type of microplastics	Grit chamber (particles/m^3)	Effluent (particles/m^3)	Increase in quantity (%)
Fibres	720–1,480	1,320–2,800	53–54
Foils	320–680	820–2,260	30–39
Granules	520–1,000	80–480	Removal 33–85
Total	1,600–3,880	2,460–5,920	65–65.5

Only granules were removed (about 33%–85%). The quantity of foils and fibers increased, despite the wastewater treatment process. In terms of material, according to the literature data, 14 polymers, mainly PE, were found at the WWTP in Germany [6]. Differences in the dominant polymer were also found when comparing raw and treated wastewater at WWTP in Denmark [26]. Acrylate, PEst, PE-PP copolymer and PP dominated in raw sewage, and acetate and PE, PEst, PE-PP copolymer and PP in treated wastewater. In turn, research from the WWTPs in California suggests that treated wastewater is not a significant source of microplastics in the environment and that these plastics are effectively removed during the skimming and settling treatment processes [27]. The majority of microplastics had a color, shape, and size similar to the blue polyethylene particles present in toothpaste.

According to the literature, the removal of plastic particles is very different. The microplastics removal efficiency is approximately 25% during the primary treatment (chemical processes) and 75% during the secondary treatment (biological) [21]. This indicates the need for additional methods that would effectively remove microplastics at the WWTP. So, what are the plastic separation methods and what is their effectiveness?

The described results of microplastics removal tests relate to the conventional WWTP, which consists of a grit chamber, the bioreactors and a secondary sedimentation tank. These systems are insufficient. Therefore, new plastic removal systems must be added to WWTP in the future. According to the literature data, the efficiency of removing this pollutant was assessed for mechanical separation, that is, disc filter (DF), rapid sand filtration (RSF), centrifuge and for the hybrid systems, that is, ballasted flocculation (BF), dissolved air flotation (DAF), UF/coagulation, biofilter and MBR. Filtration is a tertiary wastewater treatment process in WWTP enables high microplastics removal, that is, 40%–98.5% (DF) and 97.1% (RSF) [28]. During centrifuge with different speeds (5,000–10,000 rpm) and constant time (3 min) different effectiveness of nanoplastics (333 nm) removal was observed, for example, from 49% to 80% (5,000–7,000 rpm), then no change at about 80% and then 94% (10,000 rpm) [29]. The same samples were flocculated [29]. BF which is effective in the removal of small particles from water, proved less effective in the removal of nanoplastics, for example, with optimized for alum dose and after 10 min, BF achieved 77% removal. In both examples, the impurity was designated as turbidity. For microplastics and tertiary wastewater treatment with DAF, the efficiency was 95% [28]. In turn, the use of a biofilter made it possible to reduce the microplastics in the WWTP outlet by 79% (197 particles/m^3) or 89% (2.8 μg/m^3) [22].

Also, membrane processes can be used to reduce the amount of plastics released to the environment by WWTPs. Removal of PE plastics by coagulation was only 15% [21]. Increasing the efficiency of this process was achieved by adding polyacrylamide and combining it with UF. Very high removal of microplastics was observed when using MBR instead of bioreactor and secondary settling tank, that is, over 99% and even 99.9% [21,23,28]. MBR are highly efficient systems for the treatment of municipal wastewater [30].

4. Conclusion

- In all tested samples, microplastics still occur in treated wastewater in amounts that significantly increase their amount in the receiving water, despite the high efficiency of the WWTP in removing plastics.
- During the technological process, the number of microplastics increased, which may mean that plastics can be crushed into smaller parts, increasing the migration of microplastics at each subsequent stage of the wastewater treatment process, for example, 53%–54% in the case of microfibers and 30%–39% for microfoils. Greater amounts of this pollution in the effluent than in the secondary sedimentation tank may indicate the decomposition processes of organic matter at the bottom of the sedimentation tank and, consequently, the release of gases that can stick to microfoils and microfibers and carry them to the effluent.
- Effective removal of microplastics can be ensured by an additional element of the WWTP, such as filtering device for wastewater treatment. The use of this type of equipment allows almost complete elimination of microplastics from the wastewater stream. Filtration devices should be placed at the post-treatment stage, e.c. after the secondary sedimentation tank. This could prevent the increased amounts of microplastics in the effluent.
- The use of an additional wastewater treatment element opens up many possibilities, including the application of membrane techniques. While the impurities that are retained on the quick filters will be periodically washed out during their operation and diluted in water again, the use of membrane processes will allow to obtain a highly concentrated retentate, from which it will be relatively easy to separate microplastics.
- Regardless of the separation techniques used and their effectiveness, it should be remembered that the separation of microplastics is only the transfer of this pollutants

from macroplastics to microplastics and not their disposal, because microplastics are not completely biodegradable in the municipal wastewater, unlike other anthropogenic pollution.

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