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Seasonal variation and complex analysis of microplastic distribution in different WWTP treatment stages in Lithuania

Ieva Uogintė · **Sonata Pleskytė** · **Julija Pauraitė** · **Galina Lujanienė**

Abstract Industrial wastewater, domestic wastewater, and stormwater are the three entry points for microplastics (MP) in wastewater treatment plants. Extreme weather conditions, such as rising temperatures and heavy rainfall caused by climate change, can alter the rate at which MP enters wastewater treatment plants. In this study, wastewater and sludge samples from different treatment stages were collected during a 12-month sampling campaign (seasonal) to determine the efficiency of a municipal wastewater treatment plant in removing microplastic particles. MP ranging from 20 to 1000 μm were detected and classified by shape, color, size, and chemical composition. All samples contained MP particles, with concentration ranging from 1964 ± 50–2982 ± 54 MP/L in influent to 744 ± 13–1244 ± 21 MP/L in effluent and 91.1 ± 8–61.9 ± 5 MP/g in sludge; 71.6–90.1% identified particles were fragment-type with black, white, and transparent colors. Most of the microplastic particles were removed in the activated sludge tank, while the average removal rate in the wastewater treatment plant was 57%. The total concentration of MP was 27% higher in spring than in other seasons. The most common microplastic particles were polyethylene terephthalate (PET), polystyrene (PS), and polypropylene (PP). These results demonstrate the value of long-term monitoring and MP quantification, which would provide a more accurate estimate of MP pollution from wastewater treatment plants.

Keywords Microplastic · Wastewater treatment plant · Sludge · Pollution · Seasons · Complex analysis

Introduction

The growing threat of environmental disasters is one of the greatest challenges humanity is facing in the twenty-first century. In early 2012, the United Nations Environment Program (UNEP) reported that major environmental problems, such as global warming, are caused by fossil fuel use, climate change, food waste/loss, deforestation, biodiversity loss, disturbed ecosystems in coastal waters, and increasing pollution of water sources (Anon, 2012). Since the beginning of mass production in the 1950s, global plastic production has grown exponentially. Plastic is a synthetic, organic polymer with high molecular weight, usually produced by polymerization of monomers from oil, gas, coal, or by covalent solid chemical bonds (Ivleva et al., 2017). In recent decades, plastic has become one of the most important materials in our daily life and for industrial purposes. From 1950 to 2020, global plastic production has increased on average from 1.5 to 367 million tons annually (Association of Plastic Manufacturers,

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2020). An enormous amount of plastic is used in the packaging, building, construction, and automotive industries (Plastics Europe, 2018, 2019). Only 18% of plastic waste is recycled, 24% is incinerated, and the remaining 58% ends up in the natural environment (Chamas et al., 2020). Most plastic waste is found in the marine environment (Hammer et al., 2012). In the northeastern Mediterranean Sea, for example, 60–80% of the litter has been identified as plastic waste (Akarsu et al., 2020). The pervasive problem of microplastic (MP) in water sources is a global issue as it is harmful to aquatic organisms and accumulates in the human body through biological chain amplification (Cheng et al., 2021; Tang et al., 2021). Two hundred and sixty-seven different marine microorganisms have been affected by plastic waste (Moore, 2008). The number of fish, birds, and mammal species affected by plastics or MP is not known but is estimated to be in the millions. A study by Wang et al. and Banaee et al. showed that MPs can alter tissue damage, sex-specific reproductive disorders, and biochemical and immunological parameters of the blood of higher-order organisms (Banaee et al., 2019; Wang et al., 2019).

There are different types of plastics, depending on their size. Macro-plastics are considered > 5 mm, MPs < 5 mm, and nano-plastics < 0.1 μm (Tang et al., 2020). In general, MP are divided into two categories: primary and secondary. Primary MP are manufactured and used in household and industrial products such as cosmetics, personal care products, detergents, textiles, and many other everyday products (Castañeda et al., 2014). Secondary MP are released from larger plastic wastes through mechanical action, UV radiation, and microbiological degradation. For a long time, MP was considered harmless, inert particles without toxicity, but now an increasing number of studies describe the potential risks to the human body (Anbumani & Kakkar, 2018; Cox et al., 2019; Smith et al., 2018). MP may also be responsible for the increase of immune or neurodegenerative diseases. In addition, they can release hazardous additives from the matrix or serve as vectors for virulent microorganisms (Kirstein et al., 2016). In regions with intensive use of the plastics industry and a large population, there are serious environmental problems. Most of the plastic produced and consumed ends up in wastewater treatment plants and represents a further input to marine and terrestrial ecosystems.

Wastewater treatment plants can be a link between the urban environment and natural waters, such as rivers, seas, and oceans. Most wastewater treatment plants collect domestic and industrial wastewater. For example, washing clothes made of synthetic materials such as acrylic or polyester releases one million tons of synthetic particles into wastewater each year. Fifty percent of these end up in the environment (Kosuth et al., 2018). Another major impact of MP is personal care products, especially exfoliants and toothpaste: up to 4000 microbeads per 1.6 g (the average application of toothpaste) (Carr et al., 2016). Depending on the season, the weather conditions such as ice melt and/or wind also contribute significantly (Conley et al., 2019). In stormwater, MP is generated by cigarette filter waste, car tires in constant contact with road surfaces, paint from road markings, and many other sources (Fältström et al., 2021).

Wastewater treatment plants are an important input pathway for MP into the aquatic environment and increase the pollution of coastal waters (Kalčíková et al., 2017; Hidayaturrahman & Lee, 2019). For example, in China, about 9.1×10^{10} MP is released daily via domestic wastewater (Tang et al., 2021). Recent studies have shown that wastewater treatment plants can remove > 95% of MP from primary wastewater (Mintenig et al., 2017; Ziajahromi et al., 2017). However, there are still many WWTPSs with removal rates of less than 65% (Tang et al., 2020). In 2014, the Norwegian Environmental Protection Agency published a report on the rapidly increasing release of MP from wastewater treatment plants. The report highlighted that MPs enter river systems via wastewater and affect the aquatic ecosystem. Norway has estimated emissions from primary sources at about 8000 tons of microplastics (1.6 kg per capita) per year (Sundt et al., 2014). Other researchers have also pointed to the need for further investigation of wastewater treatment plants to achieve better wastewater treatment outcomes (Akarsu et al., 2020; Conley et al., 2019; Mason et al., 2016).

Several factors can influence the abundance of MP in wastewater and sewage sludge. Anthropogenic activities, physical characteristics of the river, population density, treatment efficiency, and many other factors may be responsible for higher levels in the environment (Talbot & Chang, 2022). It has been observed that the concentration of MP varies between dry and wet seasons (Cheung et al., 2019;

Stanton et al., 2020; Wang et al., 2020). The rainy season can significantly affect the surface water load of MP up to 2 mm (Xia et al., 2020). Other studies have confirmed that seasonality plays an important role, with 70–80% of the annual amount entering water bodies during the rainy season (Eo et al., 2019). However, studies do not report a significant relationship between the distribution of MP and the weather period (Cheung et al., 2019; Constant et al., 2020). Given the disparate results between seasonality and precipitation effects, further studies are needed to better understand the key differences between seasons.

Although there are no specific requirements for removing MP from wastewater, it is important to conduct targeted, well-designed, and quality-controlled investigative studies. This will allow us to understand the sources, occurrence, and efficiencies of the various treatment stages/processes, as well as the significance of the potential return to the environment from treatment wastes. The size distribution of MP is a critical factor in evaluating the treatment performance of wastewater treatment plants. There is a lack of detailed information on the size distribution of microplastics in wastewater and sewage sludge, considering a complex parameterization of MP (size, shape, color) rather than just one characteristic.

In this study, the size of MP from 20 to 1000 μm in wastewater and sewage sludge was investigated in different treatment stages in a wastewater treatment plant with aeration systems. Seasonal variation analysis and complex characterization (size, type, color) were performed to better evaluate the performance of the treatment plant. Using Fourier transform infrared spectroscopy (FTIR-ATR), we also studied the chemical composition of MP to determine its possible primary origin. In this article, the WWTP in Lithuania is used as an example to better understand how environmental and technological factors affect the distribution of MP in WWTPs.

Materials and methods

Description of the investigated plant and the sampling points

Wastewater and sludge samples were collected from a wastewater treatment plant in Lithuania, which receives wastewater from industrial and domestic activities. The WWTP consists of mechanical treatment (screens of different sizes, grit chambers, sedimentation tanks), four aeration tanks, a sludge dewatering system, and modern technology for nitrogen and phosphorus removal. The technology ensures that wastewater treatment meets current environmental standards. The design capacity of the treatment plant is 225,000 m^3 per day and y, modern sludge treatment equipment–62.1 tons of dry matter per day. The sludge is thickened in centrifuges, treated in high-pressure and high-temperature thermal hydrolysis plants, digested, dewatered in centrifuges, and dried in low-temperature dryers. A schematic representation of the main stages of the wastewater treatment plant and the sampling points can be found in Fig. 1.

Wastewater samples (5L) were taken at three different locations: at the entrance of the WWTP–influent (labeled W1), after the primary clarifier (labeled W2),
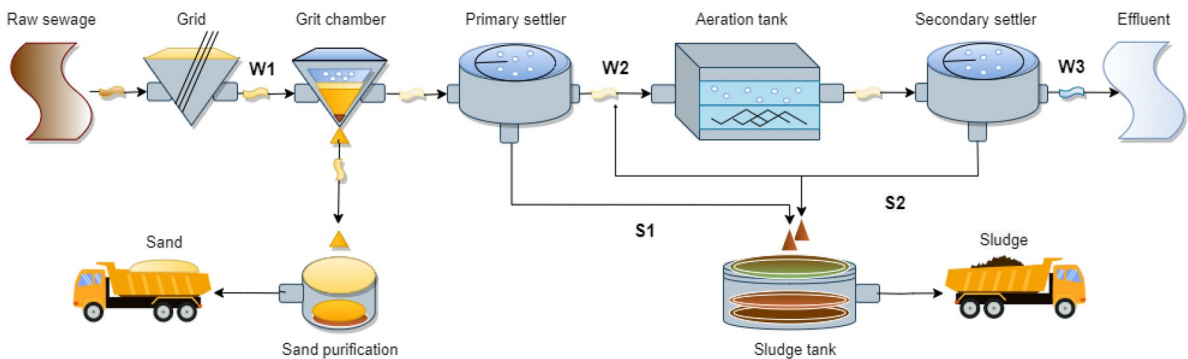

Fig. 1 Schematic diagram of the main stages of the wastewater treatment plant and the sampling points

and after the biological treatment tank at the exit of the WWTP–effluent (labeled W3). Samples of 1 kg of dewatered sewage sludge were also taken. The samples were taken in February (winter), April (spring), July (summer), and September (autumn) of the year 2021. Dark glass jars with metal stoppers were used to protect the samples from contamination. They were carefully washed several times with MiliQ water and dried at high temperatures. The wastewater and sludge samples were immediately taken to the laboratory for further processing. Over the course of a year, more than 100 samples were collected and characterized.

Extraction and characterization of MPs

The methodology for isolating and characterizing MP from wastewater and sludge samples was developed by adapting and modifying previously published work (Mason et al., 2019). It mainly consists of physical separation by filtration, chemical digestion, and counting/characterization by optical methods.

After delivery to the laboratory, MP was extracted from the wastewater samples by vacuum filtration through stainless-steel sieves of different sizes. Five sieves (> 1.02 mm, > 0.57 mm, > 0.35 mm, > 0.042 mm, and residue) with different particle sizes were obtained. Then, the MP from each sieve were transferred into pre-washed clear glass with as little water as possible for further extraction. Later, as recommended by the National Oceanic and Atmospheric Administration, the hydrogen peroxide digestion method (20 mL, 30%, 60 °C) was used to remove all organic matter and possible paper fibers. The time varied depending on the purity of the sample from 3 to 24 h. A 20 mL 1 mol/L HCl solution was then added to dissolve non-plastic (semi-synthetic or natural textiles) and inorganic materials. After this step, the samples were reconstituted in deionized water, filtered through a 1.6-μm microfiber glass filter, and dried.

As for the sludge sample, MP was first separated from the matrix using the density difference method (Li et al., 2018). A saturated $ZnCl_2$ solution (1.8 g/cm^3) was used as the separating liquid. Twenty grams of sludge and $ZnCl_2$ solution were mixed in a beaker at a ratio of 1:1, placed on the stirrer for 2 h, and then submerged for 24 h. The separation step was repeated twice to ensure that all MP had transferred from the sludge to the water. The $ZnCl_2$ solution was preferred over other common solutions such as NaCl (density 1.2 g/cm^3) to avoid the loss of particles with a higher density, e.g., polyvinyl chloride (density 1.30–1.58 g/cm^3). After separation, the supernatant was filtered and digested according to the procedure for wastewater samples.

All particles found on the filters were photographed using a digital microscope equipped with a $200 \times$ magnification lens and additional light for a better image. To avoid double MP counting, the filters were scanned in columns from the lowest to the highest edge until the entire filter was measured. This method was previously proposed by Mason (Mason et al., 2019). Only particles that met the following criteria were counted: (1) no cellular structures, (2) no additional organic material, (3) particles of uniform thickness, (4) monochromatic color, (5) only particles with a length ten times greater than the width were considered fibers.

The dimensions of MP were measured using Motic Images Plus 3.0 computer analysis software. It is important to mention that due to the chosen extraction method and technical capabilities, particles smaller than 10 μm were not detected. Although the exact size of all particles was determined, size intervals were used for further analysis. The MP determined in the wastewater and sludge samples were expressed as the number of particles per gram of dewatered sludge (MP/g) or as the number of particles per liter of wastewater (MP/L). All suspected MP were categorized by size (from 20 to 1000 μm), color (black, white transparent, red, blue, etc.), and shape (fragment, fiber, or pellet). The removal efficiency in the different stages of the wastewater treatment plant was calculated according to the following formula:

$$\text{Removal}(\%) = 100 - \frac{\text{Microplastic particles in } W_{n+1} * 100}{\text{Microplastic particles in } W_n}$$

Microplastic particle composition was determined by Fourier-transform infrared spectroscopy with attenuated total reflection (ATR-FTIR). Spectra were scanned 32 times with a resolution of 8 cm^{-1} in the range 500–4000 cm^{-1}.

SPSS 26.0 software was used for statistical analysis. Spearman rank correlation tests were performed to assess the relationships between the frequency of MP and variables likely to influence the frequency of MP (temperature, wind, and precipitation). In each

case, p 0.01 was used to determine if the results were statistically significant. t test analyses were performed to compare the mean frequency of total MP between seasons.

Contamination measures/quality control

These rules were followed to prevent contamination of the samples. Nitrile gloves and laboratory clothing made of natural materials were worn during the experiments. Only glassware was used for all experiments. All solutions were filtered through glass fiber filters before use. All surfaces were thoroughly cleaned with 70% ethanol and MilliQ water. A control was performed at each step to track contamination at the work site. Blank samples of laboratory air were also measured to determine the potential effects of airborne particles. During the experiment, the filters remained open from 9 am to 6 pm. The fiberglass filters were then examined under an optical microscope to determine if any unwanted particles or fibers were trapped. On a glass fiber filter with a diameter of 47 mm and a pore size of 10 μm, several (up to 5) particles of MP fibers were detected in the air samples within 9 h. However, their size was larger than 1 mm and did not fall within the range chosen in our study. A blank sample with distilled water contained up to 20 ± 3 particles/L.

Results and discussion

Seasonal variations in MP concentration and removal efficiency

Recent studies show that the concentration of MP in wastewater and sewage sludge varies with season and time of day (Lares et al., 2018). Weather conditions for each measurement period were examined to understand the potential relationship with MP concentrations. For each sampling period, key parameters such as air temperature (T), wind speed, wind direction, humidity (RH), and precipitation were assessed (Fig. 2). One week prior to winter sampling, wind direction was predominant from E (18.5% of all trajectories) and ENE directions (18.8%). The maximum wind speed reached 8.6 m/s, while the average one was only 3.3 m/s (standard variation ± 1.9 m/s). As expected, it was coldest in the winter period

(average, -10.5 ± 5.4 °C), and RH was the highest ($85.7 \pm 5.6\%$). One week before spring sampling, winds blew mainly from southern directions (20.0% from S and 15.8% from SSE) and on average reached 17.4 ± 7.7 m/s, which was the highest within all sampling periods. While spring weather had milder temperatures (on average 6.4 ± 5.6 °C), the humidity was highly alternating ($63.1 \pm 22.9\%$). During the summer sampling week, the prevailing wind directions were the same as in the winter period (30.6% from E and 21.0% from ENE), but wind speed was higher (10.9 ± 4.6 m/s). During the same summer period, the average temperature increased to 21.8 ± 2.8 °C, and the relative humidity remained lower ($70.8 \pm 12.7\%$). The final sampling was conducted in the autumn, and the week prior to sampling could be described as slightly cooler than summer (10.7 ± 3.9 °C) with relative humidity ($74.7 \pm 17.1\%$). During the same period, two main wind directions were observed: from the south (19.7%) and from the northwest (18.7% from NW and 15.7% from NNW). The latter was associated with higher wind speed (up to 26 m/s), while on average, it reached 11.1 ± 6.2 m/s.

In addition to the analysis of the above meteorological parameters, precipitation time was also observed for each sampling week. During winter sampling period, 41.1% of all week was snowing, and the mist was present for 7.7% of the time. During the spring, precipitation was observed 12.5% of the time and consisted of rain, snow, and wintry mix. The least precipitation was recorded during the summer and autumn sampling weeks, when it rained only 5.1% and 4.5% of the period, respectively. While the highest accumulated precipitation level was registered during the summer (22.36 mm), it was mainly linked to one thunderstorm. Meanwhile, accumulated precipitation levels in winter (1.02 mm) and spring (4.57 mm) were lower but more events of showers and snow storms were observed.

As suspected, changes in the amount of MP were observed between the different seasons (Table 1). A significantly higher number per liter in influent was observed in spring (2982 ± 54 MP/L) compared to the other seasons (2185 ± 38 MP/L in winter, 2140 ± 33 MP/L in summer, and 1964 ± 50 MP/L in autumn season).

During the winter sampling week, especially in the last 2 days before sampling, a large amount of fallen snow was noted (Fig. 2F). Due to low temperatures
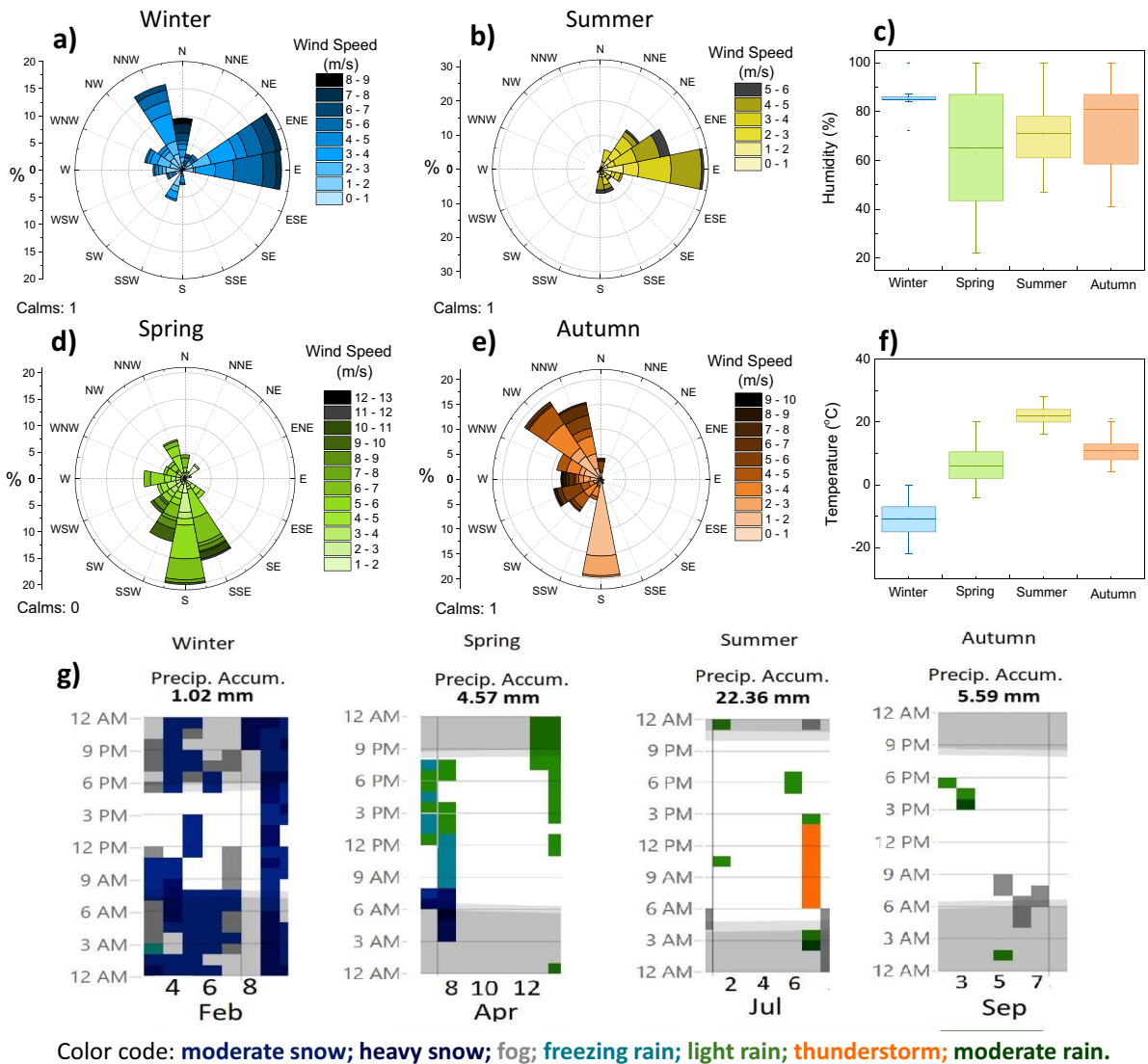
Fig. 2 Wind roses for 1-week period before each sampling during winter (**a**), spring (**d**), summer (**b**), and autumn (**e**). Box charts of humidity (**c**) and temperature (**f**) for the same time intervals. Time series of precipitation before sampling in each season (**g**)

Table 1 The number of MP per liter in each WWTP treatment stage in a different season

	Winter	Spring	Summer	Autumn
Influent	2185 ± 38 MP/L	2982 ± 54 MP/L	2140 ± 33 MP/L	1964 ± 50 MP/L
Primary treatment (sedimentation tank)	1569 ± 27 MP/L	2184 ± 35 MP/L	1700 ± 41 MP/L	1364 ± 19 MP/L
Secondary treatment (aeration tank)/effluent	877 ± 10 MP/L	1244 ± 15 MP/L	1100 ± 14 MP/L	744 ± 13 MP/L
Sludge	76.8 ± 7 MP/g	91.1 ± 8 MP/g	68.2 ± 6 MP/g	61.9 ± 5 MP/g

(as low as − 20 °C), MP cannot easily enter the waste-water treatment plant because it becomes trapped in ice or snow. Nevertheless, a relatively high amount of MP can be associated with heavy air pollution from traffic in winter (Jiang et al., 2020a, b). Traffic flow analysis has shown that car traffic in urban environment increases in winter compared to other seasons (https://portal.sisp.lt). Since car tires are considered one of the main sources, these results support the idea that many MP enter the wastewater system through them. The results show that about 27% more MP enters the wastewater treatment plant in spring, than in the other seasons. At the beginning of the week, there was still wintry weather, snow fell, and then there was a period when there was no precipitation. Two days before sampling, it rained for a long time. As indicated by the air analysis, strong winds and long-time rainfall during the spring season may have resulted in higher particle transfer. In addition, the rising temperature in spring (above 0 °C) is the cause of melting ice and snow, which contain a variety of debris, including MP. This suggests that there may be a relationship between the transitional period of the year, especially spring, and an increased number of MP (Cheung et al., 2018). Compared to spring, the number of MP decreased significantly in summer due to hot and dry weather. Only one thunderstorm event was observed 3 days before sampling. This could be decisive for the determined amount of particles in the WWTP, because the short period of time and the large amount of precipitation could wash away all microplastic particles and dilute them with a large amount of water. Also, MP particles may be influenced by thermal effects (the average temperature in July is up to 20 °C), interaction with direct sunlight, and other environmental factors (Tian et al., 2021). In addition, recent studies show that the abundance of MP in sediments and accumulation in soil are slightly higher in hot and dry seasons than in wet seasons (Xia et al., 2021). The number of MP in autumn samples

is similar to that in summer. According to the annual climatological reports, typical summer weather without big thunderstorm in Lithuania has extended into October (https://weatherspark.com/).

The results of the t test show a significant difference between the season and the frequency of MP. Table 2 shows the Spearman correlation coefficients, indicating the relationship between the concentration of MP in the wastewater entering the treatment plant and different air parameters. Accordingly, there is a negative relationship between temperature and the concentration of MP. In addition, a statistically significant negative correlation was found between humidity and the detected amount of MP. A strong positive correlation was also found between the amount of precipitation and the concentration of MP. Accordingly, more microplastics enter the wastewater treatment plant when the amount of precipitation is higher. The above statistical analyses support the authors' hypothesis that the amount of precipitation has a significant influence on the concentration of microplastics in the WWTP influent.

The number of MP per liter also decreased depending on the treatment stage. After primary treatment, it reached 1569 ± 27 MP/L in winter, 2184 ± 35 MP/L in spring, 1700 ± 41 MP/L in summer, and 1364 ± 19 MP/L in autumn. After secondary, i.e., biological treatment stage, the number of MP in wastewater was further reduced to 877 ± 10 MP/L in winter, 1244 ± 15 MP/L in spring, 1100 ± 14 MP/L in summer, and 744 ± 13 MP/L in autumn. In the sludge samples, 76.8 ± 7 MP/g was found in winter, 91.1 ± 8 MP/g in spring, 68.2 ± 6 MP/g in summer, and 61.9 ± 5 MP/g in autumn. The MP trapped in the sewage sludge provides an excellent opportunity to be recycled back into the wastewater due to environmental turbulence. In addition, if this sludge is used in landfills, it may return to the treatment plant via leachate or stormwater runoff. European WWTP sewage sludge samples from countries like Spain (165–183

Table 2 Spearman's correlation coefficient analysis

Correlation coefficient	MP amount	Temperature	Wind speed	Humidity	Precipitation
MP amount	1.00	−0.600	0.200	−0.400	1.00**
Temperature	−0.66	1.00	0.200	−0.400	−0.600
Wind speed	0.200	0.200	1.00	−0.800	0.200
Humidity	−0.400	−0.400	−0.800	1.00	0.400
Precipitation	1.00**	−0.600	0.200	0.400	1.00

** Correlation is significant at the 0.01 level

MP/g) (Edo et al., 2020), Finland (27.3 MP/g) (Lares et al., 2018), Ireland (41.96–158.35 MP/g) (Mahon et al., 2017), and Germany (1–24 MP/g) were found to contain MPs (Lassen et al., 2015). The composition of raw wastewater, local lifestyles, population, varying sampling techniques (simple or composite, sampling duration, sieve mesh size, etc.), extraction techniques (such as digestion and the use of salt for density separation), identification techniques, and sampling location are just a few of the factors that can affect the amount of MP in a WWTP. For example, the number of MP detected in the samples depends on the selected pore size of the filters. The smaller the filter size is set, the more particles are seen (Leslie et al., 2017). In this study, the amount of microplastic particles in the incoming water was lower than in many other previous studies in Europe, but higher in the outgoing water. It mostly depends on the installed WWTP treatment system. For example, according to Simon et al. (2018), the mean concentrations of MP in raw influents and treated effluents from 10 selected wastewater treatment plants in Denmark were 7216 MP/L and 54 MP/L, respectively.

With each stage of wastewater treatment, the removal efficiency of microplastics increased. Table 3 shows the overall removal efficiency (A) and the removal at each wastewater treatment stage (B) at different times of the year. After primary treatment, 28.1% of MP was removed in winter, 26.7% in spring, 20.5% in summer, and 30.5% in autumn. Large suspended solids and oils are removed from wastewater by sedimentation or air flotation during primary treatment. The removal efficiency of the WWTP increased after the second treatment stage, 44.1% of particles were removed in winter, 43.0% in spring, 35.3% in summer, and 45.3% in autumn. In the second stage, smaller inorganic particles and organic matter were removed by biological treatment. In some wastewater treatment plants, additional flocculants may be added. The overall removal efficiencies for MP in

the size range of 20 to 1000 μm were 59.8% in winter, 58.2% in spring, 48.5% in summer, and 62.1% in autumn. The average removal efficiency of this wastewater treatment plant for microplastic particles after the first treatment stage was 26.45%, after the second, 41.95%.

As can be seen from the data presented in Table 3, the treatment performance of the wastewater treatment plant is not significantly dependent on the seasons. This shows that the effectiveness of removal depends on the physical and chemical properties of the microplastic particles. However, the overall removal performance of the wastewater treatment plant may be affected by a second biological treatment stage in which organic matter is removed and a degradation process of microplastics can occur. The number of tiny microplastic particles may increase. Installation of an additional tertiary treatment stage can increase the removal efficiency up to 97%.

Size distribution of MPs

The abundance of MP in the size ranges 20–50 μm, 50–100 μm, 100–200 μm, 200–500 μm, and 500–1000 μm is shown in Fig. 3. The results are presented in the terms of the number of particle per liter and the percentage distribution of particles in a given stage to more accurately determine and explain the distribution. In addition, the MP in the sludge samples is given as the number of particles in 20 g of sludge. The largest amount was found in the size range of 50 to 100 μm. This range accounted for 32.6 to 64.4% of the total particles in the sample. The size range between 20 – 50 μm and 100 – 200 μm formed the second largest proportion: 13.9 – 17.0% and 24.6 – 27.8%, in winter, respectively; 11.6 – 23.9% and 19.3 – 28.7% in spring, 8.7 – 28.0% and 24.7 – 28.0% in summer, 5.4 – 16.4% and 24.7 – 26.3% in autumn. Fewer particles were found in the larger sizes. In the range from 200 to 500 μm, 10.6 – 17.9% MP were

Table 3 Total removal efficiency % (A) and MP removal % (B) in each WWTP treatment stage at different seasons

	Winter		Spring		Summer		Autumn	
	A	B	A	B	A	B	A	B
Primary treatment (sedimentation tank)	28.1%	28.1%	26.7%	26.7%	20.5%	20.5%	30.5%	30.5%
Secondary treatment (aeration tank)	**59.8**%	44.1%	**58.2**%	43.0%	**48.5**%	35.3%	**62.1**%	45.4%

The bold data show the cumulative efficiency of microplastic particle removal after both treatment stages in the wastewater treatment plant
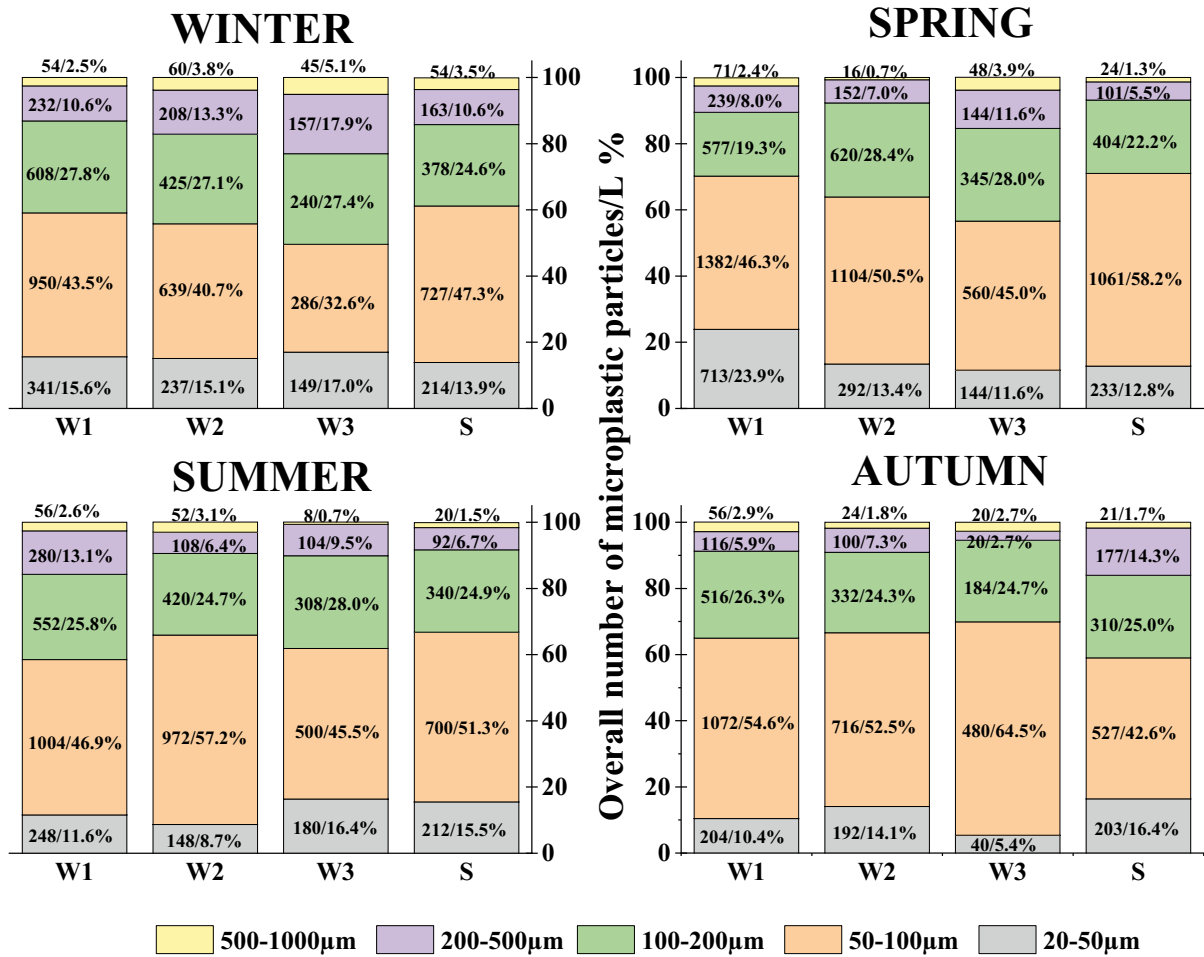
Fig. 3 Seasonal abundance and size distribution of microplastics in different WWTP treatment stages

found in winter, 7.0 – 11.6% in spring, 6.4 – 13.1% in summer, and 2.7 – 14.3% in autumn. At the same time, only a few percent larger than 500 μm were counted: 2.5 – 5.1% in winter, 0.7 – 3.9% in spring, 0.7 – 3.1% in summer, and 1.7 – 2.9% in winter. No difference in size distribution was observed between wastewater and sludge samples. Comparison of the sizes of MP in the influent (W1) and effluent (W3) at all seasons showed that the number of particles in the size ranges 20 – 50 μm, 50 – 100 μm, and 100 – 200 μm decreased by 30% after the first treatment stage of precipitation (W1 and W2) treatment step and by 20 – 50% after the second treatment stage, leaving an overall removal of about 60%. The slightly higher removal efficiency was observed for particles with size between 200 – 500 μm and 500 – 1000 μm

in summer and winter, respectively. About 60–80% of the total amount of particles were removed after the first and second stages.

An investigation of the removal efficiency as a function of size MP showed that the removal efficiency strongly depended on the number of particles in each size range. The higher the amount in a given interval, the more difficult it is to remove them in the WWTP. It was also observed that a decrease in larger particles (in the 200 – 500 μm and 500 – 1000 μm size ranges) leads to an increase in smaller particles in the 50 – 100 μm and 20 – 50 μm size ranges (thus decreasing their removal efficiency). Smaller particles may have formed either in the primary sedimentation tank due to mechanical forces or in the secondary treatment stage (aero tank) due to biological

processes and degradation of organic matter, including MP. According to studies, flocculation and gravity sedimentation are effective primary treatment methods to intercept larger particles (> 1000 m). The larger MP tend to settle to the bottom of the pond or adsorb to suspended solids and are trapped on the grid. In addition, secondary treatment has been shown to greatly reduce the amount of MP in the secondary effluent that were larger (i.e., > 500 m) (Lofty et al., 2022). Due to the high adsorption capacity of the sewage sludge, it was also more likely that MP with small particle sizes were absorbed by it and remained there (Hamidian et al., 2021). It was also found that after the aerobic process, the secondary clarifiers were where the MP removal occurred during the post-treatment process. Murphy et al. (2016) also suggested that smaller particles may remain suspended in the wastewater while larger particles sink to the bottom of the tank (Murphy et al., 2016). Since there are no specifications for the size of particles that must be determined to calculate the efficiency of MP removal in wastewater treatment plants, it is difficult to compare the results obtained with those of other researchers. Most studies found in the literature deal with sizes ranging from 100 μm, 250 μm, or 500 μm up to 5 mm (Gündoğdu et al., 2018; Jiang et al., 2020a, b).

Color distribution of MPs

The percentage color distribution of MPs is shown in Fig. 4. Black, white, transparent, brown, yellow,

and blue particles were identified during the study. Particles labeled "other" in Fig. 4 include green or red particles, which make up less than 1% of the total number of particles. The colors of MP can be used to trace their origin and identify chemical additives and potential environmental hazards. Blue fragments are obtained, for example, from water bottle caps. Black MP are usually obtained from car tires, various films, and industrial sources. The most commonly used black pigment in the manufacture of plastics is carbon black. However, when the pigment is bound to the product, it is insoluble and chemically stable (Long et al., 2013). White or transparent particles come from packaging, food containers, bags, etc. Most bright colors, such as red or green, come from synthetic materials washed in washing machines. Brightly colored polymers are produced using heavy metals such as chromium, copper, cobalt, selenium, lead, and cadmium (Razzak et al., 2022). Yellowish or brown discoloration of microplastics can be caused by weathering (Wu et al., 2018). Black (from 23.7 to 73.1%), white (from 10.8 to 47.3%), and transparent (from 6.5 to 16.4%) color particles were common in wastewater samples. Brown, yellow, and blue particles, on the other hand, accounted for a smaller proportion: 1.7 – 8.7%, 1.5 – 7.3%, and 0.8 – 8.9%, respectively. A similar color distribution was observed in the sludge samples. Different MP in color were observed in different seasons. In winter and spring, many black, white, and few blue, brown, or yellow MP were found. The results also show that most of the black particles

Fig. 4 Seasonal microplastic color variation and distribution in different WWTP treatment stages
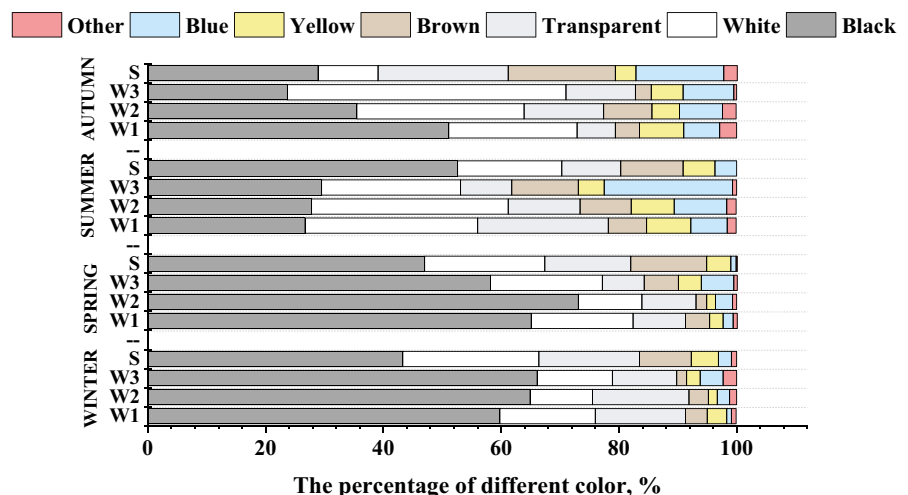
occur in the winter season (more than 60%), which is consistent with the observations presented in Part 3.1 that in winter most of the particles come from the abrasion of car tires.

In other studies, the color distribution also varied. Several authors reported that transparent color was the dominant color of MP (more than 50%) (Mason et al., 2019; Tang et al., 2020). The lower proportion of white and transparent particles in Europe is due to the European directive on single-use plastics, which aims to reduce and replace plastic bags, and food packaging with less environmentally harmful alternatives (e.g., Europe. EU). As for the removal of microplastic particles, no significant changes were observed in terms of their color. The minimal changes are due to the ability of the colored particles to interact with bacterial extracellular polymers or sludge flocs in the aeration tank, allowing them to be stored in the primary and secondary clarifiers.

Type distribution of MPs

After visual inspection, MPs were categorized by type: fragment, fiber, and pellet (Fig. 5). Fragment-type particles are considered secondary and are formed by the disintegration of larger pieces. The fiber type looks like a small thread and usually origi-nates from synthetic fabrics. Recently, it has been estimated that most fibers excreted during washing of synthetic clothing have a size of 100 to 800 µm (Hernández-Majalca et al., 2019). Round pellet-like particles are classified as primary particles because they are intentionally produced in this way. Fragment-type microplastic particles were predominant in all seasons. The proportion of this type in the wastewa-ter and sludge samples was 71.6% in winter, 86.3% in spring, 90.1% in summer, and 85.3% in autumn. Fiber particles accounted for 23.9% in winter, 11.9% in spring, 7.9% in summer, and 12.5% in autumn.
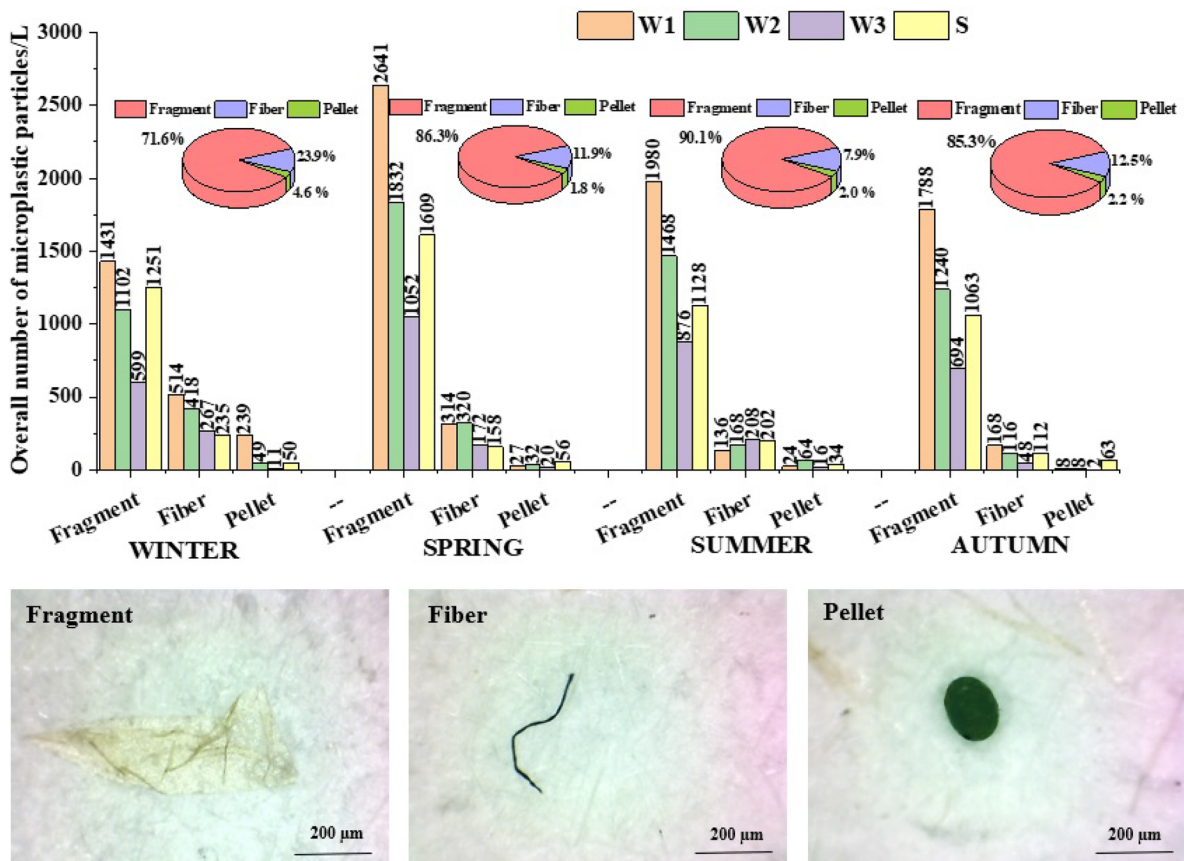

Fig. 5 Form distribution of microplastic particles in different stages of WWTP and percentage form distribution on each season

Browne et al. (2011) suggested that a higher amount of fibers would be expected in winter because people wear more clothing during the cold season (resulting in more fibers in household laundry) (Browne et al., 2011). Pellet-type particles accounted for the smallest percentage of the total MP ranging from 4.6 to 1.8%. Many other studies have found that fiber-type particles are predominant in wastewater and sludge (Eo et al., 2019; Mason et al., 2019). However, in this study, the opposite results were obtained. Our results are comparable to a study in Europe. Bayo et al. (2020) reported that 50.6% of the microplastic particles identified were of the fragment time in winter, 36.0% in spring, 59.6% in summer, and 45.1% in autumn (Bayo et al., 2020). Similar data were previously reported by Rodrigues et al. (2018) in water and sediments from a Portuguese river. The difference between the predominant types in different wastewaters may be due to population size, daily wastewater volume, wastewater characteristics, and people's habits.

The removal efficiency of each MP type in different WWTP treatment stages was also determined. Shape of the MP may affect settling behavior. About 30% of the fragment-type particles were removed in the first treatment stage at all seasons. Another 30% was removed in the second treatment stage, giving an overall removal efficiency of 60% (from 1431 to 599 MP/L in winter, from 2641 to 1052 MP/L in spring, from 1980 to 876 MP/L in summer, and from 1788 to 694 MP/L in autumn). Slightly different removal efficiencies were observed for fiber-type particles. In winter, microplastic fiber particles varied from 514 to 267 MP/L (total removal efficiency–about 48.1%). Similar removal efficiency was observed in spring (about 49.7%). The highest efficiency was achieved in autumn–after the first stage of treatment (sedimentation tank) 31%, after the second stage (biological treatment) 58.6% (total–71.4%). In summer, on the other hand, an increase in fiber particles and a negative removal efficiency between influent and effluent (−52.9%) were observed. This could be due to the ongoing processes in the WWTP where fiber particles with larger size/length are broken down into smaller particles. In this way, an increase in the number of particles would be observed.

Primary sedimentation tanks are capable of catching fragments much heavier than fibers of the same size. Large fragments (1 mm) also have lower mass and greater surface area ratio, so they are more likely to float.

Of the pellet-type MP, 25.9% were removed in spring, 33.3% in summer, 75.0% in summer, and 95.4% in winter. The high excretion rate is due to the fact that the small spherical MP can settle faster than other species (Bilgin et al., 2020).

Chemical composition of MP in wastewater treatment plant

The chemical composition of 1254 microplastic particles found in wastewater and sludge samples was determined using a ATR-FTIR spectrometer. Determination of chemical composition helps to assess the type of polymers and the origin of the source product. The following types of polymers were identified in the wastewater and sewage sludge: PE, PS, PP, PA, PET, PU, etc. (Fig. 6). The obtained results correspond to the trends in plastic production and consumption in European countries. The main uses of plastics in Europe are: PP (9.00 million tons in 2020), LDPE/HDPE (8.57 and 6.32 million tons in 2020), PVC (4.70 million tons in 2020), and PS (1.43 in million tons in 2020) (Anon, 2021).

The predominant polymer types in influent are polyethylene (27.75%), polystyrene (20.16%), and polypropylene (11.62%). These polymer types account for 59.53% of microplastic particles. Plastics most commonly used in business and industry are most abundant in wastewater. Polyethylene and polypropylene are the predominant polymer types in wastewater treatment plant effluent, accounting for 32.06% and 14.19%, respectively. Other types of microplastics (PVC, PES, acrylates, PE+PP, PET) were also detected in the WWTP effluent and accounted for 16.66% of all particles. PET and PP-containing materials were commonly used in personal care products such as toothpaste, facial cleansers, and clothing. These plastics were found in significant amounts in wastewater treatment plant effluent, suggesting that MP is caused by household wastewater pollution (Mason et al., 2019). Compared to other studies, the percentage of PE is higher (12–32%) than in China (1–6%) (Xu et al., 2020), but lower than in other European countries (e.g., 43% in Turkey (Akarsu et al., 2020)) or the USA (more than 90% (Conley et al., 2019)). PU, commonly used in synthetic textile fibers and medical devices, has also been found
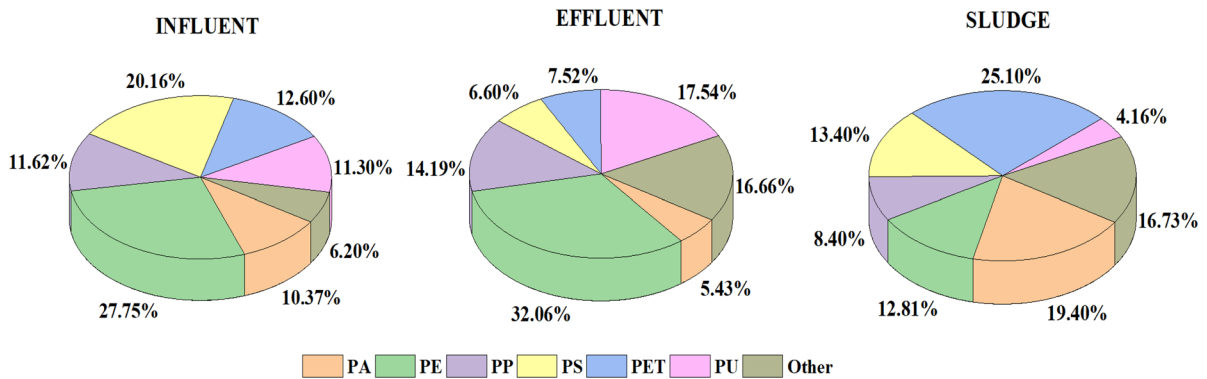
Fig. 6 Percentage of each ingredient of MPs in the WWTPs (a represented analysis of MPs with FTIR. Abbreviations: PA polyamide, PE polyethylene, PP polypropylene, PS polystyrene, PET polyethylene terephthalate, PU polyurethane)

in relatively high levels in wastewater (Magni et al., 2019). During the Covid-19 pandemic, mass production of reusable masks made of synthetic materials as a barrier to infection could also impact pollution of MP.

Particles in the influent and effluent are dominated by polymer types with density < 1.0 g/cm^3 (PE: $0.91 – 0.96$ g/cm^3, PP: $0.83 – 0.85$ g/cm^3, PS: $0.96 – 1.05$ g/cm^3). The density of these polymers determines their distribution in the stages of the wastewater treatment plant. Microplastic particles with low density are more difficult to settle during wastewater treatment, so they remain in the treatment plant and are eventually discharged into rivers together with treated wastewater. Comparing the percentage distribution of PE and PP between the influent and effluent wastewater, we find that the amount of particles of this chemical component increases, indicating a negative removal efficiency. This could be due to the fact that both PE and PP microplastic particles tend to split easily by mechanical damage. It was reported that mechanical grinding of PE and PP MPs over a 2-month period produced a significant amount of secondary MP particles, with an amount of 8.7 ± 2.5 particles per pellet for PE MP and 10.7 ± 0.7 particles/granules for PP MP (Song et al., 2017). In sludge samples, we can observe a different distribution of microplastic particles. The predominant polymer types are PET (25.10%), PA (19.40%), PS (13.40%), and various other polymers (PVC, PES, etc.) which account for 16.75% of microplastic particles. The main types of polymers identified in the sludge samples are classified as higher density plastics.

For example, the density of PET: 1.38 g/cm^3, PA: 1.13–1.35 g/cm^3, PVC: 1.38 g/cm^3, PES: 1.37 g/cm^3. Treatment with sand can effectively eliminate the particles with higher density (> 1.5 g/mL). The combination of grease skimming and pretreatment can also easily remove lower density particles, such as microbeads, which may float on the wastewater. For example, PE and PP, whose density is between 0.89 and 0.98 g/mL lower than that of water, are used to produce foams, films, and microbeads. Particles with lower density can be easily combined by skimming to form solid flakes, which can then be separated by gravity filtration. Higher density particles are also easily removed by sedimentation MP (PS, PET).

However, some studies show that the chemical composition of microplastic particles in wastewater and sewage sludge is very similar. Yang et. al. (2021) found that the percentage distribution of PP in wastewater is 33%, while in sewage sludge it is only 26% (Yang et al., 2021). The distribution of chemical composition of microplastic particles in wastewater and sewage sludge is related to human activities, technologies used in wastewater treatment plants, and the efficiency of microplastic particle removal. The prevalence of polymer types in wastewater and sewage sludge samples may differ compared to wastewater treatment plants in other regions.

Complex analysis of MPs in wastewater treatment plant

The amount of MP in the influent of wastewater treatment plants is highly dependent on several factors,

including the population served by the plants, the level of urbanization, or the industrial activity in the area (Hartline et al., 2016). For this reason, a complex analysis of microplastic particles is essential, taking into account not only their quantity but also their color and shape. Figure 7 shows an analysis for the winter season. The results show that most fragment-type particles are in smaller size ranges,

i.e., 20 – 50 μm, 50 – 100 μm, and 100 – 200 μm. Meanwhile, fiber-type particles are much larger, with predominant size ranges of 100 – 200 μm, 200 – 500 μm, and 500 – 1000 μm. In the size range of 500 to 1000 μm, a small amount of fragment-type particles were detected. No pellet-type particles were detected at all in this size range. Pellet-type particles were detected exclusively between 50 and 100 μm.
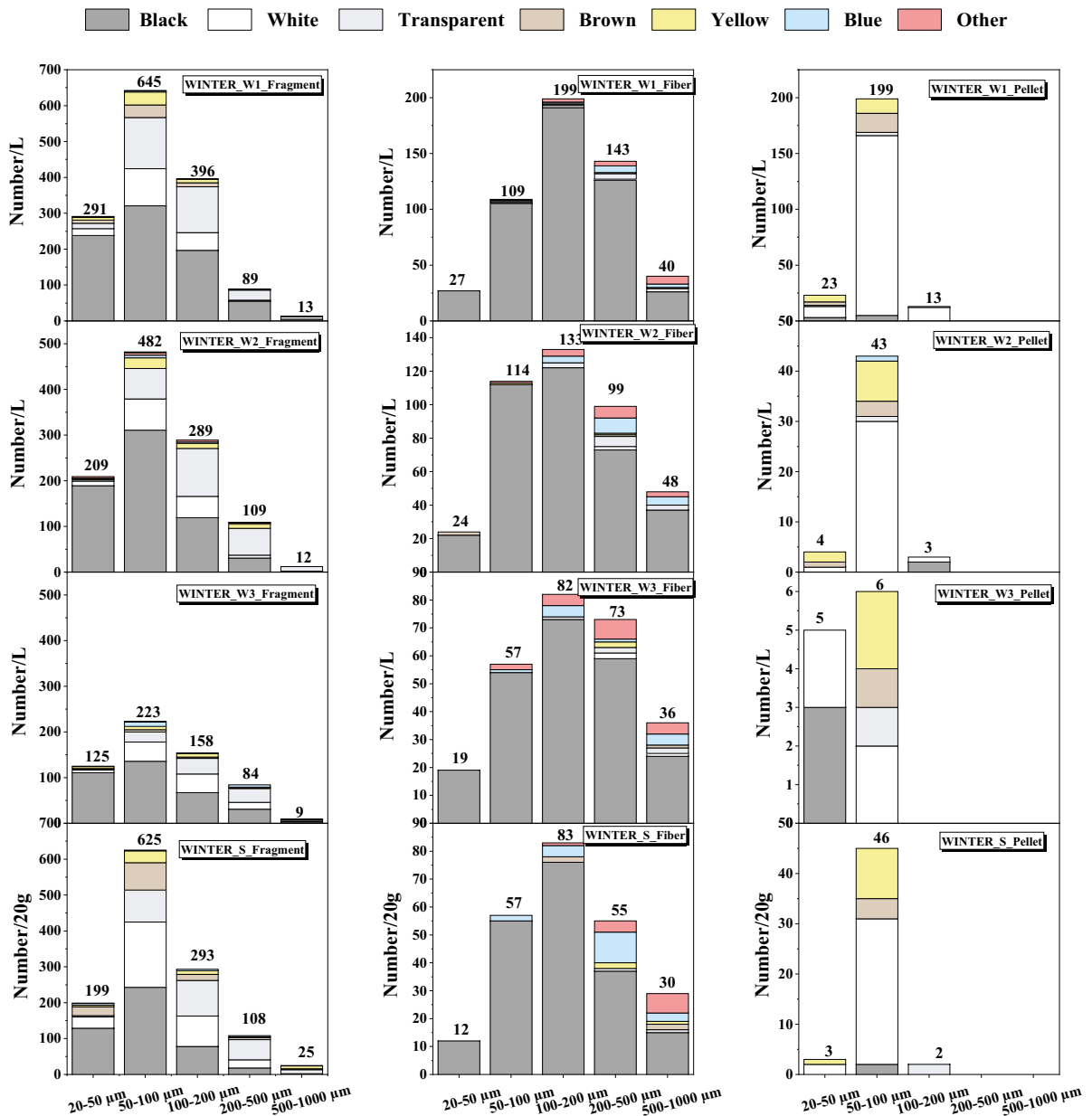

Fig. 7 Complex analysis of microplastic distribution in WWTP stages for each size range. Colors represent the color of MPs

These trends vary in the other treatment stages of the WWTP. Although black is the predominant color for fragment- and fiber-type particles, the other colors are unevenly distributed. Fragment-type particles are white, transparent, yellow, and brown, while fiber-type particles are blue and red. The granule-type particles are mainly white. The percentage of white color varies between W1 and W3 levels. The color of particles changes to yellow or brown as they pass through the various stages of the treatment plant.

MP are difficult to remove from wastewater because of their various physical and chemical characteristics (shape, size, and chemical composition). The efficiency of the fragment type particles in the size range of 20 – 50 μm, 50 – 100 μm, 100 – 200 μm, 200 – 500 μm, and 500 – 1000 μm was found to be 57.0%, 65.4%, 60.1%, 5.6%, and 30.8%, respectively. Lower removal efficiency was observed in the larger size ranges. These sizes were too small to be removed in the primary sedimentation tank but too large to be degraded in the secondary treatment stage. A similar result was obtained for fiber particles. Removal efficiencies of 29.6%, 47.7%, 58.8%, 49.0%, and 2.5% were obtained in the respective size ranges. It can be seen that the fiber-type particles are much more complicated to remove in the treatment cycle of the wastewater treatment plant. Raju et al. (2020) reported that most of the fragments are removed from the wastewater by decomposition to sludge, while the fibers remain in suspension (Raju et al., 2020).

Due to the fiber-type particle diameter of 10 –20 μm, they can easily pass through screens and filters longitudinally. Previously, Leslie et al. (2017) had shown in their experiments that fibers can pass through a membrane with a pore size of 0.08 μm (Leslie et al., 2017). Spherical polymer particles larger than 27–149 μm in diameter are most likely removed in the first sedimentation stage (Kurt et al., 2022). This is also confirmed by the results we obtained when the amount of spherical particles with a size of 50 to 100 μm decreased by about 79% after the first sedimentation step, i.e., from 199 to 43 MP/g.

Conclusion

In this work, seasonal variations and complex analysis of MP distribution in different stages of wastewater treatment plant were presented. The results showed that the amount of MP varied between seasons. Compared to other seasons, about 27% more MP enters the WWTP in spring due to strong winds, high precipitation, and ice melt. Negative relationship between temperature/humidity and the amount of MP was observed. A strong positive correlation was also found between the amount of precipitation and the concentration of MP. The largest amount of MP was found in the size range of 50 to 100 μm (32.6–64.4% of the total particles in the sample). As expected, a larger amount of fibers was found in winter. The results also show that particles of fragment type with black, white, and transparent color dominate in smaller size ranges, while fiber type (blue, red, green) is higher. The predominant polymer types in influent are PE (27.75%), PS (20.16%), and PP (11.62%), while PVC, PES, acrylates, PE+PP, PET in effluent. Many MP from our daily life end up in wastewater and are reused as sewage sludge in agriculture. This creates a closed loop. To improve the efficiency of microplastic removal, a better understanding of the transport and fate of microplastics in wastewater treatment plants is needed.

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Availability of data and materials The data used to support the findings of this study are available from the corresponding author upon request.

Declarations

Ethics approval Not applicable.

Consent to participate Not applicable.

Consent for publication All authors consent to submit this manuscript for publication.

Competing interests The authors declare no competing interests.

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