THE EFFICIENCY OF MICROPLASTIC REMOVAL BY FIVE WASTEWATER TREATMENT PLANTS WITHIN THE MONOCACY RIVER WATERSHED

by

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ABSTRACT

Wastewater treatment plants have been identified as a large contributor to the release of microplastic particles into the environment globally. The purpose of this project was to compare five different wastewater treatment facilities and their microplastic removal efficiency from treated wastewater. Another goal for this project was to determine if microplastics were entering the Monocacy River watershed in Maryland, United States. Influent, effluent, and sludge samples were collected from each facility and microplastics were extracted and microscopically examined. Microplastic particles were present in all 50 samples and microfibers under 1.0mm were most predominant. Millions of microplastic particles are entering the Monocacy River watershed from wastewater effluent and millions of microplastic particles are reentering the terrestrial environment as sludge either applied as fertilizer or deposited in landfills.

INTRODUCTION

Plastics have increasingly become a component of our lives and predominant in our waste streams. Most plastic products are single use and deposited in a landfill, being combusted, or recycled. Over 10 million tons of plastic waste were created in the U.S. between 1980s and 1990s and plastic waste continues to increase to well over 35 million tons in 2018 (EPA, 2019). Large amounts of plastic are being released into terrestrial and aquatic environments due to governmental mismanagement and societal carelessness. Some of this waste is released as microplastics, while other plastics are larger in size but eventually degrade to become microplastic particles.

Microplastics are plastic particles that are 5mm or smaller in length and can be categorized as either primary or secondary based on their origin and source. Primary microplastics are deliberately produced particles that accidentally enter the environment via manufacturing waste, stormwater drainage, and wastewater. Some examples of primary microplastics are industrial resin pellets, precursors in cosmetics, synthetic clothing, scrubs or abrasives in cleansers, and many more. Secondary microplastics originate from larger plastic items that fragment through chemical and physical processes (Alimba and Faggio 2019). Secondary microplastics are continuously breaking into smaller particles in the environment.

There are various plastic polymers in demand for production including polypropylene, polyethylene, polyvinyl chloride, polystyrene, polyurethane, and polyethylene terephthalate. These polymers are commonly used in such items as packaging, electronics, the automotive industry, houseware, lighting, and textiles (Webb et al. 2013). Since the usage and production of plastics are continuing to increase, microplastic concentrations in the environment are predicted to increase and bioaccumulate in the coming decades (Rainieri and Barranco 2019). A model created by Everaert et al. (2018) predicted that by 2100 there will be 9.6 to 48.8 particles per cubic meter floating in the ocean. Jambeck et al. (2015) estimated that 4.8 to 12.7 million tons of plastic waste enters the oceans each year and Van Sebille et al. (2015) estimated that 236,000 tons of plastic particles are currently in the world's oceans.

There are various anthropogenic sources of microplastics in the environment including stormwater, runoff, industry, agriculture, landfills, and inputs from wastewater treatment plants (WWTP) (Hitchcock and Mitrovic 2019). WWTPs have become a great interest to scientists doing research on microplastics. WWTPs receive residential and industrial wastewater and release treated effluent into local waterbodies. Wastewater initially goes through primary treatment. Screens are typically the first step in which larger debris like paper, rags, toys are removed from the water. After the screening, many WWTPs have grit chambers where minerals and rock material are removed. The last step of the primary treatment is the use of primary clarifiers, also known as sedimentation tanks. The suspended solids, sludge, settle to the bottom of the tank and removed for further treatment while the wastewater continues to the next step. Secondary treatment is used to remove dissolved organic material from the wastewater. In secondary treatment, there is an activated sludge process in which anaerobic and aerobic microorganisms consume the organic material. After the organic material is removed, water quality parameters are tested ensure they are within regulation. For tertiary treatment, they use chemical and physical processes to purify the wastewater further with filtration and disinfection (EPA 1998).

Liquid municipal WWTP effluent has been identified as a top contributor of microplastics entering the aquatic environment, while sludge also contains large amounts of microplastics (Edo et al. 2020). Subsequently, sludge is commonly applied as fertilizer for agriculture, which may enter the broader environment through leachate or runoff (Ngo et al. 2019). In Europe and the United States, it is estimated that 63,000 to 430,000 and 44,000 to 300,000 tons of microplastics, respectively, could be added to agricultural soils annually (Scalenghe 2018).

A study conducted by Lares et al. (2018) examined the occurrence of different plastic polymers in WWTP samples and determined that polyester and polyethylene terephthalates were the most common. They made up about 79.1% of microplastics collected from WWTP samples and they were mostly fibers and fragments. Lares et al. (2018) also determined that polyethylene made up about 11.4% of the microplastics collected and were either fragments or microbeads. Polyamide (nylon) fibers made up about 3.7% of microplastics collected. The remaining 5.8% of microplastics collected were negligible due to all the organic material making it difficult to analyze with the FTIR spectroscopy.

In the United States, there are varying estimations of how many microplastics are entering through WWTPs. Rochman et al. (2015) estimated an input of 13 billion particles per day and Mason et al. (2016) estimated 8 billion particles per day in the U.S. (Li et al. 2018). Luo et al. (2019) determined that microplastic concentrations increase downstream from WWTPs in freshwater systems when they compared different types of water bodies for microplastic pollution. Also, residential washing machines have been noted to be a prominent microfiber source in wastewater, which can input more than 1900 microplastics per item of clothing per wash cycle (Browne et al. 2011).

Most published microplastic studies have taken place in estuarine and marine environments. There is a small percentage of studies involving freshwater environments and an even smaller percentage in the United States. WWTPs discharge into many freshwater bodies around the globe. Facilities all vary in treatment processes, technological advances, and size. Some facilities have larger treatment capacities, and they treat a large volume of wastewater each day from multiple residential and industrial areas, while some WTTPs only treat wastewater from small townships. Facilities are upgraded at different times with different technologies based on the facility's current infrastructure, its location and/or influent rate. All these differences may have an impact on the amount of microplastics being released from each WWTP. There is a need for more research among WWTPs and what treatment processes are most effective at removing microplastic particles from wastewater and sludge. Doing so will help encourage facilities to upgrade their systems and/or technology for removing microplastics.

There are many WWTPs in central Maryland, several of which discharge into the Monocacy River watershed. The Monocacy River starts near in south-central Pennsylvania and travels 58 miles south through central Maryland between Carroll and Frederick County borders until it empties into the Potomac River. A map of the Monocacy River watershed and its WWTPs was created using ArcGIS 10.7 is shown in Figure 1. Water quality in the Monocacy River impacts the Chesapeake Bay. One of those impacts could be from microplastics. I investigated five WWTPs that discharge into the Monocacy River watershed. These WWTPs vary in the treatment processes they employ. The facilities being compared in this study are: Emmitsburg WWTP, Taneytown WWTP, Thurmont WWTP, City of Frederick WWTP, and Ballenger-McKinney WWTP. The goal of this study was to determine if the treatment processes at these five WWTPs along the Monocacy River are removing microplastics from the wastewater effectively or if they are being predominantly discharged into the watershed.

I hypothesized:

1. The biomembrane technology used at the Ballenger-McKinney WWTP will remove microplastics most efficiently from the wastewater due to its fine pore size of 0.04 microns.
2. The City of Frederick and Ballenger-McKinney WWTPs receive the most volume of wastewater and industrial waste, which may lead to more primary microplastic particles being incorporated into the wastewater versus the Emmitsburg, Taneytown, and Thurmont WWTPS that receive less industrial wastewater.
3. Microplastics under 1mm are not being removed by the wastewater treatments that lack a biomembrane filter.

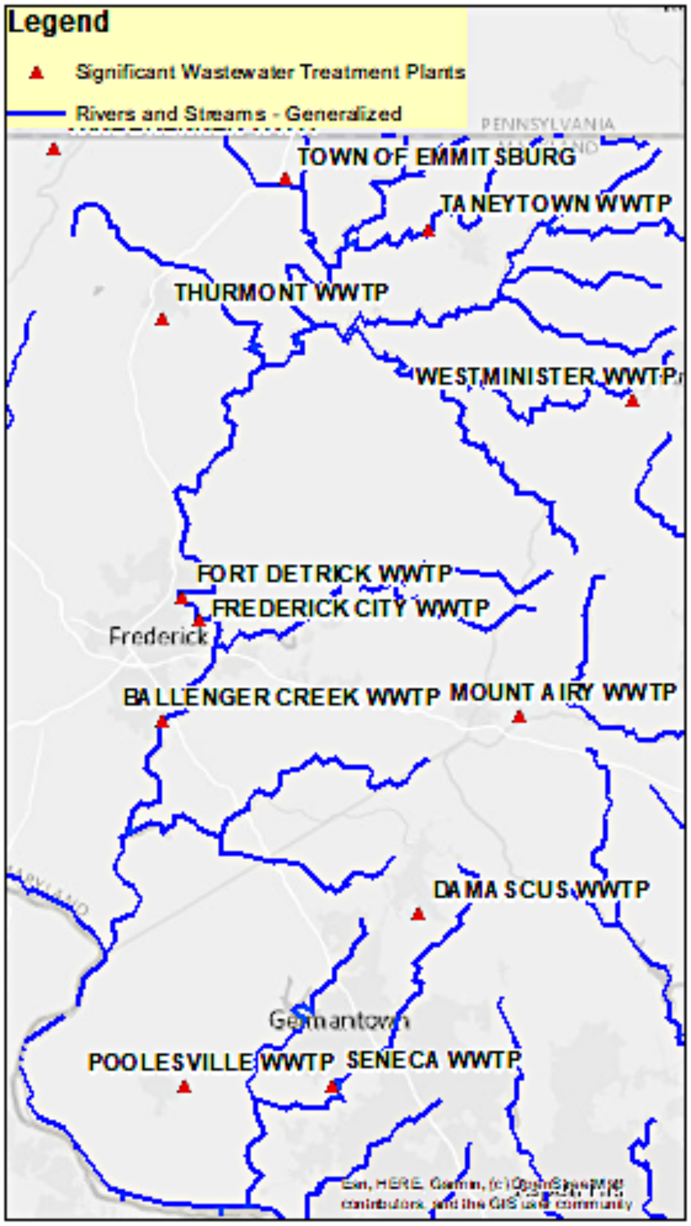

Figure 1. Map of the Monocacy River watershed and significant WWTPs within it in the state of Maryland. Made with ArcGIS 10.7 software.

<u>OBJECTIVES:</u>

The objectives of my work are four-fold.

i. Characterize the different WWTP facilities and technology used to treat wastewater in a step-by-step fashion.

ii. Determine if microplastics are being discharged into the Monocacy River watershed through WWTP effluent samples.
iii. Determine the efficiency of each treatment plant in removing total microplastics from wastewater by comparing influent to effluent concentrations.
iv. Quantify concentrations, morphological types, and sizes of microplastics in each WWTP to determine what microplastics are being removed versus those not being removed.

MATERIALS AND METHODS

Facility Comparison

Each facility was toured with a staff member and the details of the treatment processes in each plant was recorded. Table 1 compares the different characteristics of each of the five WWTPs used for this project.

Table 1. Comparison among the five WWTPs being examined for microplastic removal.

	Emmitsburg	Taneytown	Thurmont	Ballenger-McKinney	City of Frederick
Average Influent Vol (gallons per day):	~430,000	~740,000	~1 million	~15 million	~3 to 6 million
Max. Capacity (gallons per day):	750,000	1.1 million	3.5 million	22 million	40 million
Main Treatment Characteristic:	Biolac Treatment, 2 ° Treatment	Sequencing Batch Reactors (SBR), 2° Treatment	Enhanced Nutrient Removal, 3° Treatment	Biomembrane Filter, 3° Treatment	Nutrient Removal Reactors, 2° Treatment
Discharges into:	Toms Creek	Piney Creek	Hunting Creek	Monocacy River	Monocacy River

The Emmitsburg municipal WWTP is capable of processing 430,000 gallons of influent per day using a Biolac treatment process. A Biolac pond biologically treats wastewater using extended aeration, activated sludge treatment that is simple and cost-effective. This facility receives wastewater from the town of Emmitsburg and can hold 750,000 up to gallons at any one time. The treatment process is shown in Figure 2. This facility discharges into Toms Creek, which discharges into the Monocacy River. The plant's dewatered sludge is a class B biosolid that has detectable pathogens, which means the sludge cannot be used as fertilizer and is taken to a nearby landfill (Fissel, personal communication).

Figure 2. An aerial image of the Emmitsburg WWTP. Major steps in the treatment process are labeled and numbered as sequential steps (Google Earth).

The Taneytown municipal WWTP is a 740,000-gallons per day facility that employs sequencing batch reactors (SBR). SBRs are an activated sludge treatment process with influent tanks treating wastewater and effluent tanks releasing treated water. The maximum capacity of this facility is 1.1 million gallons. Its treatment process is shown in Figure 3. This WWTP discharges into Piney Creek in Carroll County and then enters the Monocacy River. The sludge is taken by a third part company called Denali Water Solutions to provide fertilizer for farming (Raab, personal communication).

Figure 3. An aerial image of Taneytown WWTP. Major steps in the treatment process are labeled and numbered as sequential steps (Google Earth).

The Thurmont municipal WWTP treats a 1,000,000 gallons of wastewater per day using enhanced nutrient removal technology. Enhanced nutrient removal process removes total nitrogen to levels as low as 3mg/L and total phosphorus to 0.3mg/L or less (Freed 2007). The facility receives wastewater from households, industries, and both the Cunningham Falls State Park and Catoctin Mountain National Park. The maximum capacity of this facility is 3.5 million gallons. Their treatment process is shown in Figure 4. This WWTP discharges into Hunting Creek, which is a class 3 trout stream, but is only marginal habitat for trout. Being a class 3 trout stream, the requirements for effluent nutrients, temperature, and pH at this WWTP are more restricted. The dewatered sludge is transported to a Pennsylvania landfill (Eyler, personal communication).

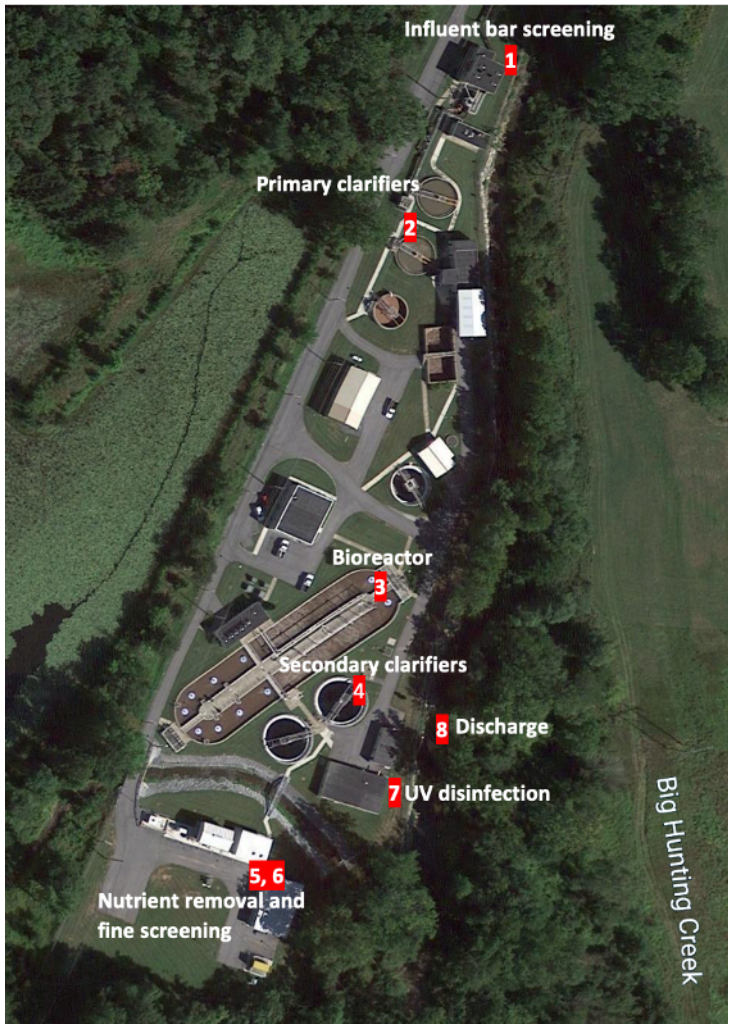

Figure 4. An aerial image of Thurmont WWTP. Major steps in the treatment process are labeled and numbered as sequential steps (Google Earth).

The Ballenger-McKinney WWTP is a 15-million gallons per day facility that receives wastewater from residences and industries in Frederick, MD. This facility is much larger than the other four WWTPs, with a maximum capacity of 22 million gallons. Their treatment process is shown in Figure 5. This WWTP discharges directly into the Monocacy River. The sludge from this WWTP is used for fertilizer in Frederick County but some batches are sent to the landfill if there are detectable amounts of pathogens in them. This plant is different from the rest because it uses a ZeeWeed 500 membrane to produce high quality effluent with a 0.04 micron filter size (McClanathan, personal communication).

Figure 5. An aerial image of Ballenger-McKinney WWTP. Major steps in the treatment process are labeled and numbered as sequential steps (Google Earth).

The City of Frederick WWTP receives wastewater from four sewer lines in the City of Frederick locality, including one from Walkersville, two from Frederick City proper, and one from the Frederick airport. In total the plant's inputs is 3 to 6 million gallons per day. The maximum capacity of the facility is 40 million gallons. The facility also uses enhanced nutrient removal with secondary treatment, which is shown in Figure 6. This WWTP discharges directly into the Monocacy River. The sludge from this WWTP is applied as fertilizer to farms in PA and a few in Frederick County (Fiery, personal communication).

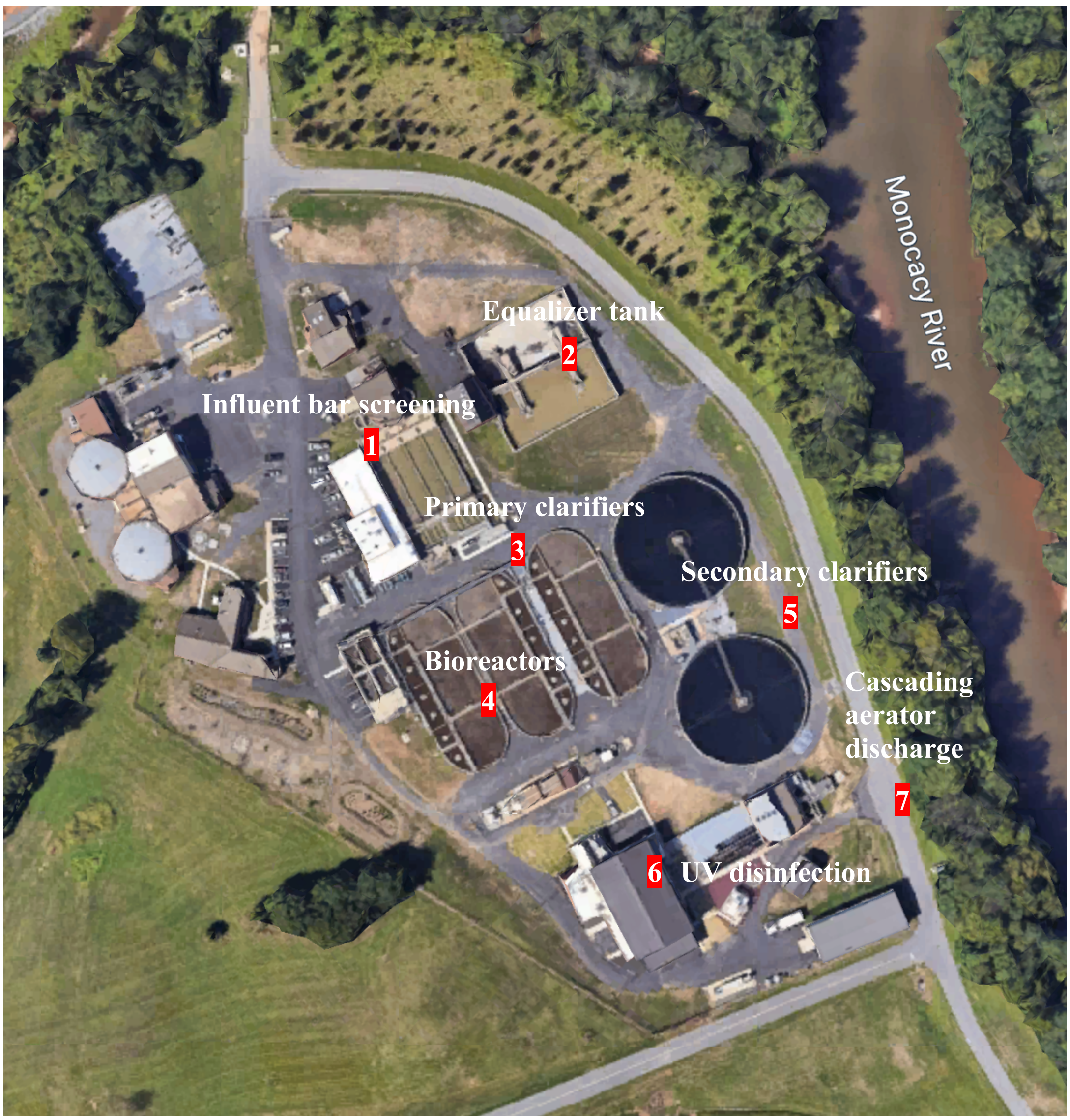

Figure 6. The City of Frederick WWTP process. Major steps in the treatment process are labeled and numbered as sequential steps (Google Earth).

Field Sampling

Three influent and three effluent wastewater samples were taken once and collected in the summertime at each wastewater treatment facility by staff members. The samples were collected all in the same day to minimize differences in weather. The amount of precipitation prior to sampling, flow rate, and estimated amount of grit and sludge was noted on the sampling day. Buckets were used to collect samples by WWTPs' staff and then placed into 250mL sterile containers from each of the WWTPs. Sludge was collected by hand and put into two 150 mL containers. The sludge being collected is their final waste product; other waste that is taken out during screenings is taken to nearby landfills.

Microplastic Extraction

Samples were placed into 250mL flasks and dried at 90 °C until all moisture was removed. To remove organic material, 10mL of 10% KOH was added for every 1g of dried sample. The samples were then placed in an incubator at 60°C for 24hrs or until all organic material was removed. Once all organic material was removed from the sample and appeared as cloudy, white color, a density separation took place. In the density separation, 5M NaCl was used because it has a higher density than water allowing for most microplastic polymers to float to the top. The top layer of water was filtered through a 0.22µm Nylon filter and microscopically examined for microplastics.

Quantifying Microplastics

I categorized microplastics by type and approximate size during microscopic examination; a copy of the datasheet used is shown Appendix 1. Particle morphotype (fibers, pellets, beads, foams, or films) is an important characteristic to determine what particles are being released most frequently. Approximate length of each plastic particle was measured with a transparent ruler during the microscopic examination to see what size range is most effectively removed during wastewater treatment. Photos were taken of all microplastics found.

Quality Assurance

Deionized water samples were used to determine if there was laboratory contamination during the microplastic extraction procedure. Filters were left out in the laboratory to determine if there was any air-borne plastic contaminants within the laboratory. There were techniques used to mitigate laboratory contamination including wearing gloves at all times, avoiding plasticware as much as possible, using aluminum foil to seal glassware, working in a fume-hood at all times to prevent air contamination, and washing glassware repeatedly with multiple rinse buckets and laboratory soap. I also wore an orange cotton shirt that was easily recognizable to determine if there is handling contamination from myself.

Three 250mL deionized water samples were spiked each with 50 microplastic particles with various types and sizes. These samples were run through the extraction procedure and microscopically examined to determine the recovery rate and whether there was a possibility of biased data.

Statistical Analysis Plan

Descriptive statistics were calculated to compare each facility. A two-way ANOVA was run to compare the mean microplastic removal from influent, and release in effluent and sludge samples of the five WWTPs. In doing so, the two-way ANOVA would determine which facility is most efficient. A t-test was used to determine if there was a significant difference among the influent microplastic concentration vs. effluent microplastic concentration for each WWTP. Descriptive statistics were also used to characterize the concentrations and types of microplastics not being removed by treatment. IBM SPSS Statistics software was used for statistical analysis.

Since the sample size for this research project has been determined by resource and time availability, a sensitivity power analysis was conducted, in which the effect size is estimated given the sample size. The anticipated effect size was determined to be 0.37 for a sample size of 40 (30 samples being wastewater and 10 being sludge).

RESULTS

Quality Assurance

The three filters that had deionized water filtered through them had an average of 3 ± 1 microplastic particles per filter membrane. The three filter membranes that were laid out to determine air contamination had an average 0.67 ± 0.6 microplastic particles per filter membrane. There is slight laboratory and handling contamination that must be taken into account when analyzing the results of the WWTP samples. The three spiked samples, done within the laboratory to measure the efficiency of the microplastic extraction methodology, had an overall recovery rate of 87%. Microfibers were recovered at 91% efficiency, while microfragments were recovered at 86% efficiency and microbeads were recovered at 83%. The recovery rate determined should be noted when analyzing the results too. Spiked microbeads and microfragments were most prone to being lost when being transferred into the vacuum filtration system, either by sticking to the beaker walls or to the walls of the Buchner funnel after rinsing with deionized water.

Microplastic Removal Efficiency

A two-way ANOVA was performed to compare the mean microplastic concentrations in influent, effluent, and sludge samples taken from the five WWTPs, which is shown in Table 2. There was a significant difference found between the numbers of microplastic particles found from each of the five WWTP. There is a significant difference in the number of microplastic particles found between influent and effluent samples.

Table 2. Results of a two-way ANOVA to compare the mean microplastic concentration in influent, effluent, and sludge samples from the five WWTPs.

Tests of Between-Subjects Effects			
Dependent Variable: Microplastics Concentration			
Source	*df*	*F*	*Sig.*
Corrected Model	9	8.574	0.794
Intercept	1	184.288	0.902
WWTP	4	0.738	0.129
Specimen Type	1	71.212	0.781
WWTP * Specimen Type	4	0.751	0.039
Error	20		
Total	30		
Corrected Total	29		

An independent samples T-test was performed to compare the influent microplastic concentration versus effluent microplastic concentration for all WWTPs; the results are shown in Table 3. There is a significant difference between the microplastic concentration of influent and effluent wastewater samples ($p<0.0001$). The interaction plot displays the difference between the influent and effluent microplastic concentration among the five WWTPs used for this study, which is shown in Figure 7. The Emmitsburg, Taneytown, and City of Frederick WWTPs have similar slopes in the interaction plot, meaning their removal rates are either the same or very similar. The Thurmont WWTP had a similar slope as well, but the microplastic concentrations were higher. Ballenger-McKinney WWTP had a steeper slope in the interaction plot, meaning their removal rate was higher. The average microplastic particle concentrations for influent, effluent, and sludge for each WWTP are summarized in Table 4. Microplastic particles are present in influent, effluent, and sludge samples from all five WWTPs. Due to their presence in effluent wastewater, microplastic particles are being discharged and entering Monocacy River watershed.

Table 3. Results of an independent samples T test to compare influent microplastic concentrations vs. effluent microplastic concentrations.

Independent Samples Test								
					t-test for Equality of Means		95% Confidence interval of the Difference	
		t	df	Sig. (2-tailed)	Mean Difference	Std. Error Difference	Lower	Upper
Microplastic Concentration	Equal variances not assumed	8.765	21.044	0.000	10.733	1.225	8.187	13.280

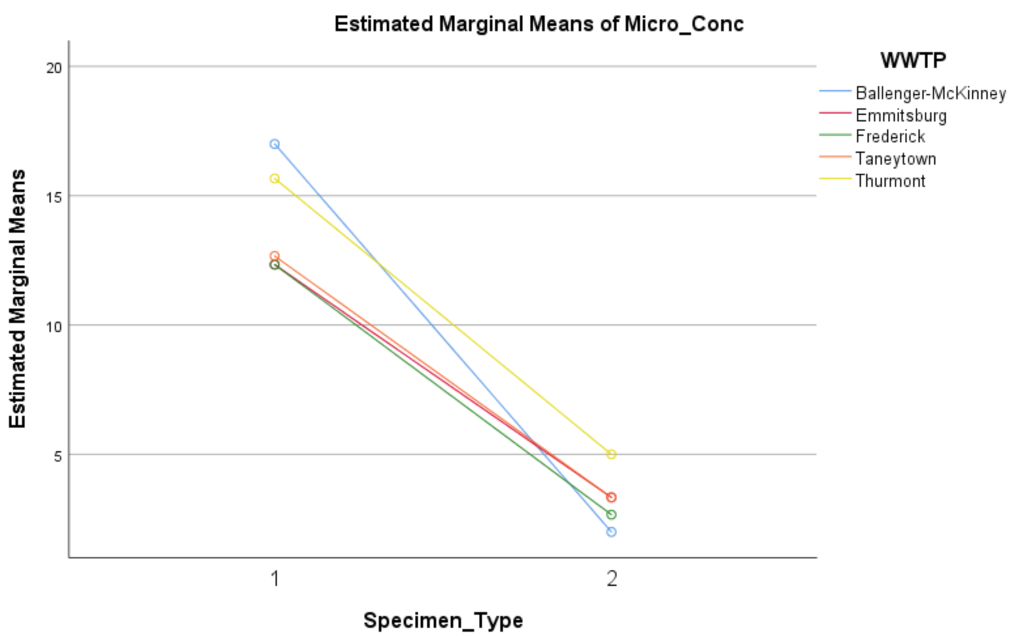

Figure 7. An interaction plot testing the difference among influent and effluent microplastic concentrations of the five WWTPs.

Table 4. Average microplastic concentrations (±stdev) in the influent, effluent, and sludge samples among the five WWTPs.

WWTP	***Average concentration of microplastic particles in influent (particles/250mL) (±stdev)***	***Average concentration of microplastic particles in effluent (particles/250mL) (±stdev)***	***Average concentration of microplastic particles in sludge (particles/5g) (±stdev)***	***Average microplastic particles not accounted for***
Emmitsburg	12.3 (±3.1)	3.3 (±1.2)	4.5 (±3.5)	4.5
Taneytown	12.7 (±1.2)	3.3 (±0.6)	4.0 (±2.8)	5.4
Thurmont	15.7 (±4.9)	5.0 (±4.4)	3.5 (±3.5)	7.2
City of Frederick	12.3 (±5.7)	8.0 (±2.1)	4.5 (±3.5)	-0.2
Ballenger-McKinney	17.0 (±5.3)	2.0 (±1.0)	6.5 (±3.5)	8.5

The Ballenger-Mckinney WWTP had the most microplastic particles within its influent wastewater, with an average of 17 ± 5.3 microplastic particles per 250mL of wastewater. The Ballenger-Mckinney WWTP receives its wastewater from many industrial facilities compared to the other four facilities. However, the Ballenger-Mckinney has the least amount of microplastic particles in its effluent, with an average of 2 ± 1 microplastic particles per 250mL of effluent. Their microplastic removal rate is estimated to be 88% and the estimated amount of microplastics being discharged into the Monocacy River watershed is 2.3×10^8particles per day using their flow rate of 7.649 million gallons per day (mgd). Liu et al. 2021 determined that tertiary WWTPs were able to remove on average 85 to 90% of microplastic particles from wastewater. Ballenger-McKinney falls within that range of other global tertiary WWTPs. Microplastic particles are also leaving the Ballenger-McKinney facility in the sludge. It is estimated that approximately 1.7×10^8 particles per day are in this facility's sludge. The sludge is either used as fertilizer or deposited in a nearby landfill. Microplastic particles are released back into the aquatic and terrestrial environments by the Ballenger-McKinney WWTP.

The city of Frederick WWTP was found to have an average concentration of 12.3 ± 5.7 microplastic particles per 250mL of influent, 8 ± 2.1 microplastic particles per 250mL of effluent and 4.5 ± 3.5 microplastic particles per 5g of sludge. The City of Frederick WWTP had the most microplastic particles release in their effluent out of the other five WWTPs and enter the Monocacy River. The treatment process at this WWTP is not removing microplastic particles as efficiently as other WWTPs with a removal rate estimated at 65%. Liu et al. 2021 determined that secondary WWTPs were able to remove on average 66 to 75% of microplastic particles from wastewater. So, the city of Frederick WWTP falls lower than the average secondary WWTP of microplastic removal. The estimated amount of microplastics being discharged into the Monocacy River watershed is $9.1x10^8$particles per day using their flow rate of 7.5mgd. The estimated amount of microplastics in their sludge is $4.8x10^8$ particles per day.

The Thurmont WWTP had an average microplastic concentration of 15.7 ± 4.9 particles per 250mL in the influent, 5 ± 4.4 microplastic particles per 250mL in the effluent and 3.5 ± 3.5 microplastic particles per 5g of sludge. The treatment process at this WWTP is also not removing microplastic particles as efficiently as other WWTPs with a removal rate estimated at 68% but falls within the range of 66 to 75% at microplastic removal in secondary WWTPs (Liu et al. 2021). The estimated amount of microplastics being discharged into the Monocacy River watershed is $6.0x10^7$particles per day using their flow rate of 0.798mgd. The estimated amount of microplastics in their sludge is $1.3x10^8$ particles per day.

The Taneytown WWTP had an average concentration of 12.7 ± 1.2 microplastic particles per 250mL in the influent, 3.3 ± 0.6 microplastic particles per 250mL in the effluent, and 4 ± 2.8 microplastic particles per 5g of sludge. The treatment process at this WWTP is removing microplastic particles more effectively than the Emmitsburg, Thurmont, and City of Frederick facilities. The estimated removal rate is 74% and falls within the range Liu et al. 2021 found of 66 to 75% at microplastic removal in secondary WWTPs. The estimated amount of microplastics being discharged into the Monocacy River watershed is 3.6×10^7 particles per day using their average treated volume of 0.786mgd. The estimated amount of microplastics in their sludge is 1.1×10^8 particles per day.

The Emmitsburg WWTP had an average concentration of 12.3 ± 3.1 microplastic particles per 250mL in the influent, 3.3 ± 1.2 microplastic particles per 250mL in the effluent, and 4.5 ± 3.5 microplastic particles per 5g of sludge. The estimated removal rate for this facility is 73% and falls within the range of 66 to 75% at microplastic removal in secondary WWTPs (Liu et al. 2021). The estimated amount of microplastics being discharged into the Monocacy River watershed is 3.3×10^7particles per day using their average treated volume of 0.728mgd. The estimated amount of microplastics in their sludge is 9.9×10^7 particles per day.

Type Distribution

Microfibers were the most common type of microplastic particles identified, followed by microfragments, and then microbeads. Eleven samples contained only microfibers and two samples did not contain any microfibers. Figure 8 shows selected images of microplastic particles found in some wastewater samples. The total sum of microfibers identified were 211 particles, while the total sum of microfragments and microbeads were 66 and 28 respectively. Figure 9 summarizes the types of microplastics in each WWTP sample.

Microfragments and microbeads were most commonly found in influent and sludge samples, meaning that the treatment processes are removing them from wastewater but are still leaving the plant in the form of sludge. Effluent wastewater samples indicated that microfibers were the most common type still present after treatment.

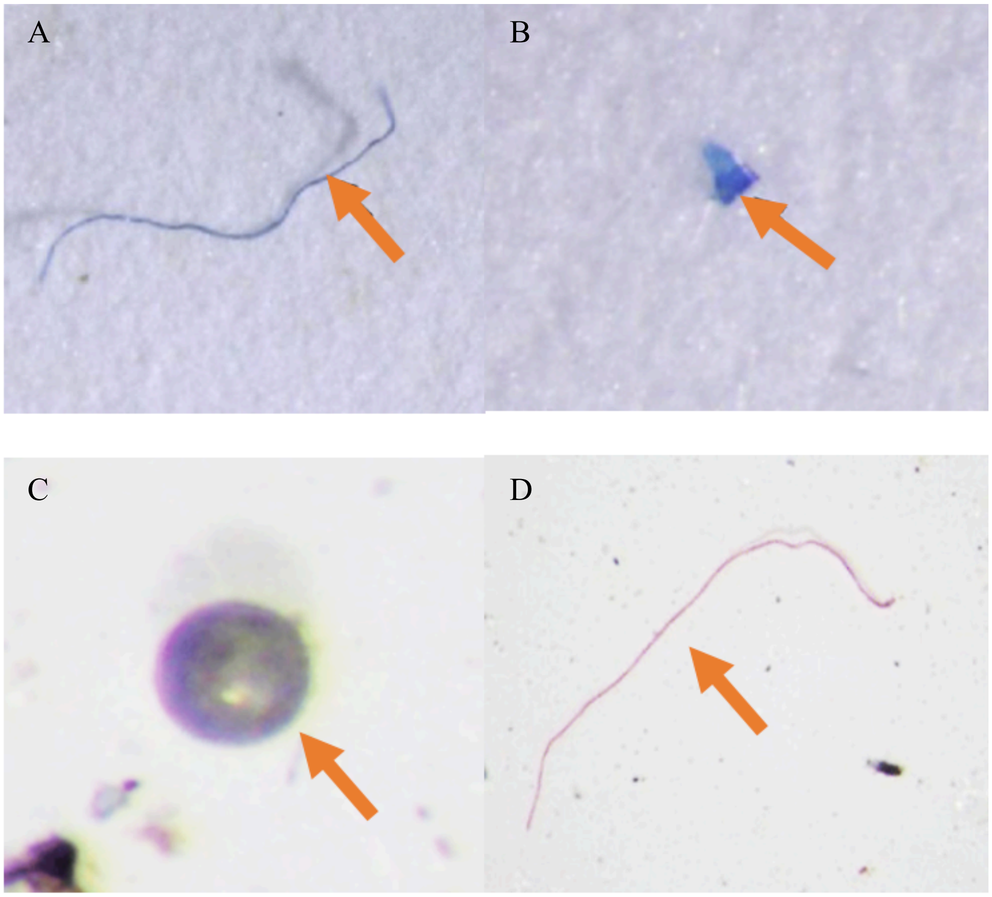

Figure 8. Photographs of microplastic particles that were extracted from wastewater samples. A) is a blue microfiber from a Thurmont effluent sample, B) a blue microfragment from a Ballenger-McKinney influent sample, C) a microbead from a Ballenger-McKinney influent sample, D) a red microfiber from a Taneytown effluent sample.

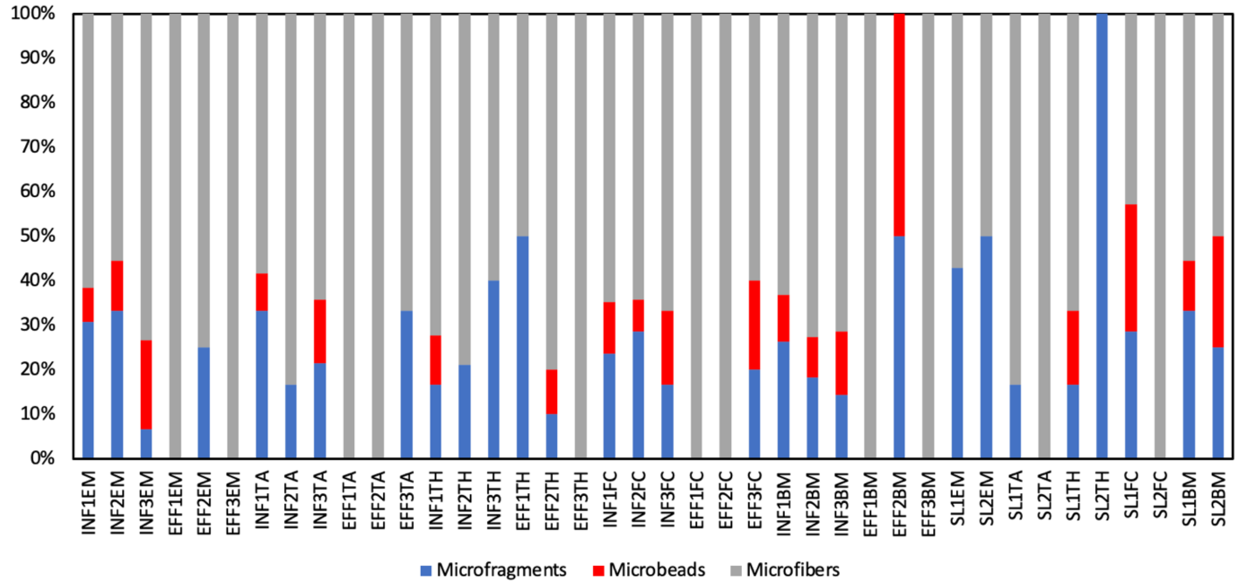

Figure 9. Microplastic type distribution among the 40 samples. INF1EM stands for influent, sample 1, from Emmitsburg. EFF1EM stands for effluent, sample 1, from Emmitsburg. SL1EM stands for sludge, sample 1, from Emmitsburg.

Size Distribution

Microplastic particles that are under 1.0mm were most commonly found in the WWTP samples. The size distribution of 5.0mm and larger were the least present size distribution found in the effluent samples. The microplastic size distribution for all 50 samples is shown in Figure 10. Majority of the larger microplastics are being removed from the wastewater once treated, but they are still present in the sludge. Microplastic particles still present in the effluent samples were under 5.0mm in length, but most of them were less than 1.0mm long.

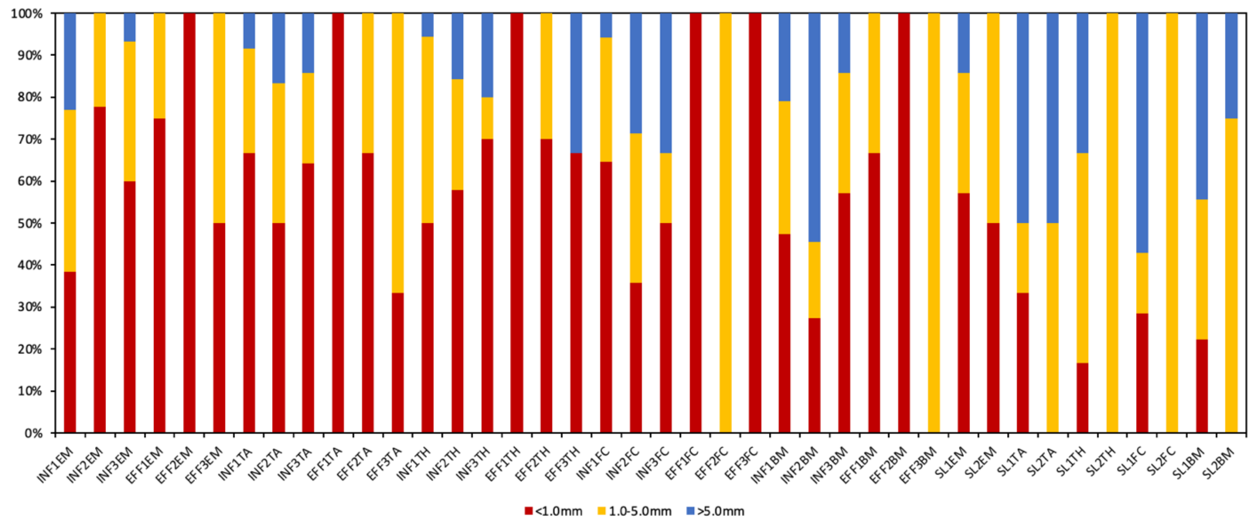

Figure 10. Microplastic size distribution among the 40 samples. INF1EM stands for influent, sample 1, from Emmitsburg. EFF1EM stands for effluent, sample 1, from Emmitsburg. SL1EM stands for sludge, sample 1, from Emmitsburg.

Limitations

Examining influent, effluent, and sludge samples from multiple WWTPs for microplastic particles has shown differences among each plant. Estimations were made to determine approximately how many microplastic particles are being released from WWTPs. The morphotype and size that are most commonly found in effluent samples were noted. Sludge was also found to contain a large portion of microplastics. This project is a great initial study done on WWTPs in the Monocacy River watershed. However, there were some limitations to this study that should be taken into account for future research.

Wastewater and sludge samples were only collected on one day. Collecting samples on multiple times may lead to a better understanding and comparison of microplastic concentrations in wastewater. Also sampling multiple times would lead to the possibility of different weather patterns, which may affect the contaminants and pollutants in the wastewater.

Another limitation to this study was the methodology of how wastewater was collected. City of Frederick was the only WWTP to use an automatic sampler, the other four WWTPs either used a dipper multiple times or used the containers to collect the wastewater samples. Using a dipper or container to collect wastewater only grabs what is at or near the surface of the tank. The tanks may or may not have a homogeneous mixture with pollutant distribution in the wastewater.

A third limitation to this study was that sampling was conducted shortly after a 0.29inch rainstorm. This rainstorm allowed stormwater to transport more pollutants to the WWTPs and it increases the flow rate of each WWTP (McMahan).

Greater resolution in microplastics could have been discerned by sampling at each step in the treatment process of the five WWTPs. This was not done due to limitations of resources and time. If each treatment step was sampled, the possibility of narrowing down which treatment step is removing majority of the microplastic particles from wastewater would have been helpful in determining why a particular WWTP is doing better or worse than others.

Sampling the waterbodies above and below the WWTPs for microplastics would help solidify that these WWTPs are discharging significant microplastics into the Monocacy River watershed; however, this was not done.

Microscopic examination was the only methodology used to quantify microplastic particles in samples. It is not as reliable as other methods including FTIR or Raman spectroscopy.

A final methodological limitation is that microplastic particles under 0.22μm would pass through the filter membrane used to collect microplastics from the density separated sample. Microplastics are known to break down and further fragment in the environment, including during treatment at WWTPs. Particle sizes could be down into the nano scale and be passed through the filter membrane.

DISCUSSION

WWTPs in the Monocacy River watershed are not removing 90% of microplastic particles from the wastewater. More than 10% of microplastics from the influent wastewater are present in effluent wastewater (Iyare et al. 2020). Ballenger-McKinney is the most technological advanced WWTP looked at in this study. With the use of multiple screenings and a membrane filter, Ballenger-McKinney had the highest removal rate of microplastics in this study. The mesh size of both the fine screening and membrane filter prevents most microplastic particles ranging 0.04µm to 6mm from being discharged by the plant. It has been noted in Iyares et al. 2020 that tertiary wastewater treatment plants can range from 82.1 to 99.9% microplastic removal. However, microplastic particles of all sizes are present in sludge samples. Currently there is no treatment done at Ballenger-McKinney to remove them from sludge. The sludge is used as fertilizer at local Frederick County farms or it is deposited at the local landfill.

The City of Frederick is a large WWTP that does not use technological advance methods for treatment. It is recommended that this plant is in need of upgrades the most because it was the least efficient at microplastic removal. There is no policies or regulations in place for wastewater treatment plants to remove microplastic particles from wastewater in the United States. It only removes about 65% of microplastics from the wastewater, with majority remaining in the sludge final product. The enhanced nutrient removal process works well for treating wastewater with high levels of nutrients but is inefficient at removing microplastics. A fine screening or membrane filter would be useful in this situation to further filter out particles from the wastewater that are under 5mm.

The Thurmont WWTP also uses enhanced nutrient removal as their main treatment process. It removed approximately 68% of microplastics from the wastewater. However, Thurmont uses a fine screening unlike the City of Frederick WWTP. The fine screening is able to filter out particles between 1.5 to 5mm (EPA 2003). Membrane filter is still recommended to further filter microplastic particles that are under 1mm in size to increase the efficiency of the Thurmont WWTP.

Emmitsburg WWTP is 73% efficient at removing microplastics from the wastewater by using a Biolac pond system and secondary treatment. Biolac ponds use aerated and unaerated techniques to remove nutrients from wastewater and sludge. This facility also recirculates solids into the Biolac pond to dilute the organic content (Pennsylvania Department of Environmental Protection). Due to the recirculation, microplastics may stay in the recycled activated sludge resulting in fewer microplastics from leaving the plant.

The Taneytown WWTP is unique and has sequencing batch reactors with magnetite to increase the rate of sedimentation. Taneytown is 74% efficient at removing microplastics, which is the second highest efficiency. Using magnetism may have negatively charged plastic polymers like polypropylene, polyester, and polystyrene separated from the wastewater. Zhang et al. 2020 found that polystyrene nanoplastic particles has a strong electrostatic attraction to magnetite, which is a common plastic polymer.

The average number of microplastic particles coming into the WWTPs is more than the average number of microplastic particles leaving the plants for the Emmitsburg, Taneytown, Thurmont, and Ballenger-McKinney WWTPs. Microplastic particles can end up being filtered out by the fine screenings and membrane filters, which these particles are not accounted for. Those particles would be deposited in the landfill along with the other waste that was filtered out. Microplastics can also be breaking down into the nanoscale by the treatment processes. Those particles would not be separated out by the methods used in this study. The 0.22μm

Nylon filter would allow nanoplastics pass through during the extraction phase. The average number of microplastic particles coming into the WWTPs is less than the average number of microplastic particles leaving the plant for the City of Frederick WWTP. More microplastic particles can end up in the discharge after treatment due to fragmentation. Fragmentation can cause a particle to be broken up into numerous smaller particles, which can add to the number of particles being discharged from a plant.

Microfibers under 1.0mm in length were the most prominent microplastic particles found in effluent wastewater samples. The shape and size of microfibers make their removable more difficult because they can pass through filters perpendicularly so there is less surface area (Poerio et al. 2019). Large inputs of microfibers come from domestic washing machines that can discharge 20 to 240lbs per 100,000 people (Harrison 2017). However, microplastic concentrations in wastewater is dependent on population density, industries, and urbanization (Raju et al. 2020). Membrane filters and membrane bioreactors are added to WWTPs to reach higher microplastic removal rates (Poerio et al. 2019). Leslie et al. 2017 reported that microfibers were passing through a membrane with a pore size of 0.08μm. Even though Ballenger-McKinney membrane has smaller pores, microplastics are not being filtered out of the wastewater and more research is needed into membrane filters and bioreactors.

Microplastics entering the Monocacy River watershed may be consumed fish and other aquatic organisms. Many health issues can occur to these organisms including internal bleeding, abrasions and blockages in the digestive tract that can lead to starvation. Microplastics are also known to absorb contaminants from waterbodies that can release harmful toxins into the body of organisms. These toxins can affect endocrine systems, reproductive systems, immune systems. Bioaccumulation and biomagnification of the particles and toxins will disrupt the Monocacy

River watershed and Chesapeake Bay watershed food chains. Eventually this will lead to humans ingesting microplastics through consumption of aquatic organisms, which has already been documented (Harrison 2017).

Deposited microplastics on agricultural cropland is a huge concern too for human health. With some of the WWTPs have farmers deposit sludge containing microplastics to fertilize the soil, microplastics are reentering the terrestrial environment. Corradini et al. 2019 found microplastic concentrations higher in agricultural soils that were receiving sludge applications. It has been documented that particles 50nm in size can penetrate crop roots, but it is possible for particles that are 2000nm to penetrate crop roots too. An Italian study looked into crops sold at local stands versus supermarkets and found that local stands tended to have higher levels of microplastics on crops (Toussaint 2020). As stated before, the endocrine, reproductive, and immune systems can be compromised in organisms, including humans, if the plastic concentrations continue to increase within the body.

More research is needed to determine which treatment step is removing majority of microplastics from wastewater in the five WWTPs to understand what specific treatment steps are removing microplastics. More research on what microplastic removal techniques may be added to WWTPs and their permits to make them more efficient is another possible track. Iyare et al. 2020 determined that primary treatment can remove about 72% of microplastic particles from wastewater, while secondary treatment can remove a further 16%. The study also determined that tertiary treatment could remove about 94% of microplastic particles from wastewater (Iyare et al. 2020). Examining sites above and below the five WWTPs would be a helpful future project to determine the microplastic concentrations present in the Monocacy River watershed and how WWTPs affect the ecosystem. Sampling cropland soil, corn and soybeans in Maryland would be another helpful project to see if they are being affected by microplastics would also be an interesting future project because corn and soybeans are two major crops Maryland produces.

APPENDIX 1

Datasheet to categorize microplastic characteristics.

Sample ID: ______	<1.0mm	1.0-5.0mm	>5.0mm
Microbead			
Microfiber			
Microfragment			
Microfilm			
Microfoam			
Total			

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