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Occurrence and Removal of Microplastics in Tertiary Wastewater Treatment Plants: A Case Study of Three Plants in Zhengzhou, China

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Abstract: Microplastics have been widely detected in wastewater treatment plants, but there is still a significant dearth of research data on the removal efficiency of microplastics in such plants. The present study focused on three wastewater treatment plants situated in Zhengzhou, China. On-site sampling and Raman spectrum detection techniques were employed to identify microplastics in both wastewater and sludge samples, while the removal efficiency of microplastics was quantified for each plant. Results showed that the abundance of microplastics in influent exhibited ranging from 147.5 ± 2.6 to 288.8 ± 11.8 n/L, while the range in sludge samples was from 12,024.7 ± 1737.0 n/kg$_{dw}$ to 20,818.4 ± 5662.0 n/kg$_{dw}$. The removal efficiencies of microplastics in the three WWTPs ranged from 76.2% to 91.2%. The primary components of microplastics were generally identified as fibers ranging in size from 10 to 100 μm. The samples collectively exhibited a total of seven distinct colors, with the predominant proportion being transparent. Polypropylene was the polymer type with the highest proportion. The sludge in WWTPs plays a pivotal role in the accumulation of MPs from wastewater bodies, necessitating increased attention toward its proper disposal in future endeavors.

Keywords: microplastics; wastewater treatment plant; tertiary treatment process; removal efficacy

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1. Introduction

The widespread application of plastic products in the fields of agriculture, construction, and manufacturing is primarily due to their remarkable resistance to corrosion, stability, and cost-effectiveness [1]. Extensive utilization and persistent non-degradability of plastic waste have given rise to a myriad of environmental and ecological predicaments [2]. According to the United Nations Environment Programme, an annual influx of over 8 million tons of plastic is observed in the ocean [3]. Plastic waste in the environment is subject to the influence of various environmental factors, including wind, sunlight, and water flow. This leads to a process wherein it gradually disintegrates from a larger solid into smaller solid particles. Previous studies have introduced the academic concept of microplastics (MPs), which are defined as plastics with a size smaller than 5 mm [4]. Due to the minute size of MPs, they can easily serve as carriers for other pollutants, facilitating their migration and transportation across various environmental media such as water and air; this phenomenon exacerbates the extent of environmental pollution [5]. The investigation of MPs in aquatic environments originated from the study of marine ecosystems, where numerous scholars had conducted extensive research on the spatial distribution patterns, transport mechanisms, and ecotoxicological impacts of MPs in oceans, yielding a substantial body of scientific findings [6,7]. MPs are also widely presented in freshwater and were frequently detected in rivers, lakes, reservoirs, and groundwater [8]. The World Health Organization report highlights significant variations in the abundance of MPs in freshwater, which can differ by several orders of magnitude [9]. However, the studies on its pollution characteristics and pollution mechanisms have not been sufficiently in-depth, and our understanding of the cycle process between human society and natural water remains limited [10].

The wastewater treatment plants (WWTPs), serving as a crucial receptor of urban wastewater, also act as a significant contributor to the natural water system. For traditional WWTPs, nitrogen, phosphorus, and organic matter are the mainly targeted pollutants to be removed [11,12]. Therefore, the predominant treatment processes employed for them primarily utilize biological methodologies. Given that MPs are predominantly derived from anthropogenic activities, serving as a crucial interface between human society and natural water systems, WWPTs hold significant potential in facilitating their recycling process [13,14]. Similarly, some scholars have conducted extensive research on the migration, transportation, distribution, and removal efficacy of MPs in WWTPs [15,16]. The sedimentation after the biological process in WWTPs, as a crucial unit for the removal of MPs, exhibits a significant removal efficiency ranging from 78% to 99% [17,18]. The removal rate, while not insignificant, still allowed for a substantial annual influx of MPs into surface water through effluent from WWTPs. In contrast to drinking-water treatment plants, secondary WWTPs typically lack coagulation units, thereby relying primarily on the encapsulation and adsorption of activated sludge for the removal of MPs in WWTPs. Previous studies have demonstrated that the incorporation of coagulation units into secondary wastewater treatment plants can significantly enhance the removal efficiency of MPs, with laboratory experiments achieving a maximum removal rate of 99.4% [19,20].

In order to comply with more stringent environmental regulations, numerous WWTPs have undergone upgrades and renovations, predominantly incorporating tertiary treatment processes. Physical and chemical methods have emerged as the most prevalent approaches in this regard [21,22]. However, the detection and analysis methods for emerging contaminants such as Pharmaceuticals and Personal Care Products (PPCPs), Endocrine Disruptors (EDs), Antibiotics Resistance Genes (ARGs), Persistent Organic Pollutants (POPs), MPs, etc., have only been mentioned in recent years, and our knowledge of the removal efficiency and mechanism of WWTPs for these contaminants is limited [23,24]. The removal of MPs in WWTPs is significantly influenced by various factors, including the types of treatment processes, the contribution of rainwater, the daily treatment capacity, and the operation mode, as indicated by previous studies [25,26]. Currently, a standardized approach for sampling and detection methods of MPs is lacking, while the availability of comprehensive data on the removal efficacy of MPs in operational WWTPs is currently limited. Therefore, in order to establish a standardized protocol for the collection and detection of MPs in the future, as well as to accumulate fundamental research data on the baseline levels of MPs in natural water environments, further extensive investigation is imperative and holds significant reference value.

The present study investigates the occurrence and removal efficiency of MPs in three urban WWTPs located in Zhengzhou city. We analyzed the abundance, sizes, morphologies, colors, and polymer types of MPs in wastewater and sludge samples from WWTPs, while also discussing the factors influencing MP removal within these WWTPs. We explore the disparities in MPs among different WWTPs and with a recognition of sludge as an aggregation environment for the accumulation of MPs. Concurrently, we acknowledge the limitations of this study and aspire for its data to serve as a scientific reference for deliberations on MP removal in WWTPs, thereby offering scientific strategies to mitigate MP discharge into surface water.

2. Materials and Methods

2.1. Field Sample Collection

The research subject of this study consisted of three urban WWTPs located in Zhengzhou, which were equipped with tertiary treatment processes. For the purpose of maintaining confidentiality, the WWTPs were not given a specific name and were referred to as WWTP-A, WWTP-B, and WWTP-C, which had treatment scales of 600,000 m^3/d, 200,000 m^3/d, and

100,000 m^3/d, respectively. The sampling points (Figure 1) for wastewater samples were strategically placed at the influent (IN), secondary treatment process outlet (STPO), and effluent (OUT). The experimental specimen was prepared by collecting 2.5 L of wastewater using a stainless steel bucket and then storing it in a glass bottle [27]. In order to improve the accuracy of the experiment, three samples of wastewater (7.5 L in three bottles) were collected from each sampling point. For sludge samples, the wet sludge samples (1 kg) from each WWTP, were carefully enclosed in aluminum foil bags for the purpose of detecting MPs. The samples were subsequently subjected to further processing in the laboratory.

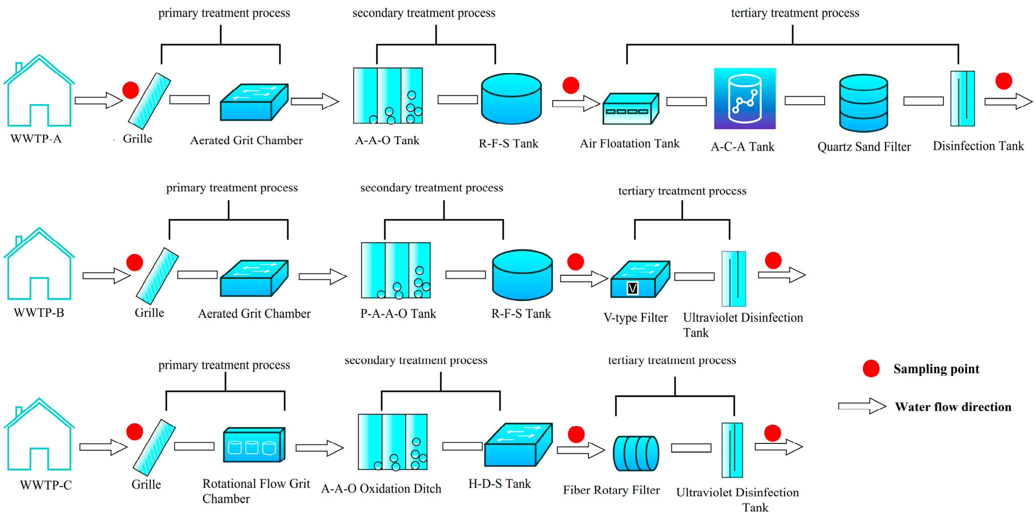

Figure 1. Location of wastewater sampling points in different WWTPs (A-A-O: Anaerobic-Anoxic-Oxic; P-A-A-O: Preanoxic-Anaerobic-Anoxic-Oxic; R-F-S: Radial-Flow-Sedimentation; H-D-S: High-Density-Sedimentation; A-C-A: Activated-Coke-Adsorption).

2.2. Extraction of MPs

The methods employed for the extraction of MPs in wastewater and sludge samples in this investigation were based on previous research, with specific steps adjusted to accommodate the unique conditions of the samples [28,29]. Firstly, the large pieces of garbage in wastewater were picked out with stainless steel tweezers, and then wastewater samples were subjected to a slow filtration process using a stainless steel wire sieve (10 µm of pore size). The intercepted substances on the sieve were subsequently rinsed with 50 mL of ultra-pure water and collected in a 200 mL glass beaker. The liquid was treated with 50 mL of 30% H_2O_2 (Sinopharm Group Chemical reagent Co., LTD, Shanghai, China) and incubated in a water bath at 50 °C for 12 h to facilitate the degradation of organic compounds. Furthermore, an excess amount of NaCl (Sinopharm Group Chemical reagent Co., LTD, Shanghai, China) was introduced into the glass beaker to create a supersaturated solution and subsequently allowed to stand undisturbed for a duration of 12 h to facilitate the process of density stratification. The liquid was then passed through a water ring distillation and filtration unit using a glass fiber mesh membrane (0.45 µm of pore size) (Delvstlab Co., LTD, Haining, Zhejiang, China). Then, the filtered membranes were stored in glass culture dishes. A total of three independent experiments were conducted on water samples at each sampling point.

For each WWTP, 100 g of each sludge sample was placed into a 1 L glass beaker respectively, followed by the simultaneous addition of saturated sodium chloride solution (600 mL). The solution was completely homogenized by agitating it with a rotational speed of 200 r/min using a magnetic stirrer for 30 min. Then, these beakers were subsequently immersed in an ultrasonic water bath for a duration of 20 min, aiming to enhance the subsequent separation efficiency of MPs from the sludge. After a 12 h standing period, the liquid was filtered by a stainless steel wire sieve (10 µm of pore size). Subsequently, the intercepted materials were thoroughly rinsed using ultra-pure water (50 mL) and collected in a 500 mL glass beaker. The above steps were repeated three times for each individual sludge sample to ensure the optimal extraction efficiency of MPs. The beaker was then supplemented with 150 mL of 30% H_2O_2, followed by incubation in a water bath at 50 °C for 12 h to facilitate the digestion of organic compounds. The liquid was ultimately subjected to a water ring distillation and filtration unit using a glass fiber mesh membrane (0.45 μm of pore size), and the membranes were subsequently collected in glass culture dishes. Similarly to the wastewater samples, the sludge samples were also subjected to three separate experiments for each WWTP. Additionally, 200 g of wet sludge samples from each WWTP underwent oven drying at 105 °C for 48 h in order to determine their moisture content.

2.3. Identification Method of MPs

The filtered membranes were positioned on the objective table of an optical microscope (Motic BA310Digital, Motic, Xiamen, China), and a camera was connected to a computer for data transmission. Image collection and processing software (Motic Images Plus 3.0 ML, Motic, Xiamen, China) was employed for the collection of suspected objects on the filtered membranes. The quantity, dimensions, shapes, and hues of the suspected objects were documented and captured in photographs. The orientation information of suspected objects was confirmed by the mesh on the filtered membranes. The types of MP polymers were identified through a confocal Raman microscopy spectrometer (CRMS, gora-Lite, Ideaoptics, Shanghai, China). In this investigation, the parameters of this spectrometer were configured to cover a wave number range spanning from 60 to 3500 cm^{-1}, achieving a high resolution below 5 cm^{-1}, and employing laser excitation at a wavelength of 785 nm. Generally, the suspected objects present on the filtered membranes in wastewater samples were individually detected, and the obtained results of spectral data were subsequently analyzed using the data processing software in the CRMS system (gora. Dawn v1.0, Ideaoptics, Shanghai, China). Whereas, in light of the excessive amount of suspected objects in filtered membranes of sludge samples, instead of individually testing all suspected objects, we adopted a methodology based on previous studies and conducted a random selection of the objects for examination [30]. In this study, we randomly chose 100 particles in each filtered membrane to ensure maximum diversity in the light of dimensions, shapes, and hues. The purpose of doing so was to ensure the scientific rigor of experimental results. By referencing a previous study, we conducted a comparative analysis between the detected spectral data and the standard spectral data, establishing that it corresponded to a specific polymer when the similarity exceeded 70% [31]. The quantification and subsequent analysis of particles identified as MPs were conducted upon the completion of all tests.

2.4. Quality Control

The stainless steel buckets, glass bottles, and aluminum foil bags utilized for sample collection and storage underwent thorough cleaning with ultra-pure water followed by drying in the oven prior to usage. In the laboratory setting, researchers donned cotton lab coats and disposable latex gloves for each experiment. Throughout the experimental procedure, all equipment was covered with aluminum foil during intervals between operational steps to prevent airborne MPs contamination of the samples. Additionally, MP extraction was conducted within a fume hood test bench.

2.5. Data Analysis

In this research, Microsoft Office 2010 software was used for data statistics and calculation, SPSS Statistics 24 software was used for a one-way analysis of variance (ANOVA), and Origin 2018 was applied to draw the pictures.

3. Results and Discussion

3.1. Abundance of the MPs

MPs were observed in all collected samples, including water and sludge samples, with varying levels of abundance (Figure 2a). The detailed data can be found in Tables S1 and S2. The IN wastewater displayed a wide range of abundance from 147.5 ± 2.6 to 288.8 ± 11.8 n/L, and the OUT wastewater appeared a range of abundance from 20.8 ± 4.6 to 68.8 ± 2.0 n/L. The abundance of MPs in IN wastewater in this study was consistent with the conclusions drawn from some previous research [32,33]. In this study, a one-way analysis of variance (ANOVA) was employed to assess the abundance of MPs in each WWTP, and the results showed that there were significant differences among the three WWTPs. The p-values were 0.000037 (IN), 0.000202 (STPO), and 0.000008 (OUT), respectively. It is evident that MPs predominantly originate from human activities, which appears to contribute significantly to the observed variations in MPs' abundance across different WWTPs. The WWTP-A and WWTP-B are situated within the urban core area of Zhengzhou City, catering to a substantial population with high levels of human activity. Conversely, WWTP-C is located on the outskirts of Zhengzhou, exhibiting a significantly lower abundance of MPs in IN wastewater compared to the other two plants. Researchers have conducted a comprehensive analysis of the occurrence of MPs in various WWTPs, revealing substantial variations in the abundance of MPs within the influent, even exhibiting significant differences spanning several orders of magnitude [34]. For traditional WWTPs, nitrogen, phosphorus, and organic matter are the mainly targeted pollutants to be removed, and the concentration of pollutants in the wastewater remains relatively stable during the initial design phase of these WWTPs [11]. However, there are currently no specific regulations regarding the permissible levels of MPs in wastewater treatment standards. The substantial fluctuations in the abundance of MPs in influent can lead to varying pollution load conditions for WWTPs during operational processes [35]. In the future, it is imperative to establish a standardized protocol for detecting and evaluating MPs in WWTPs.

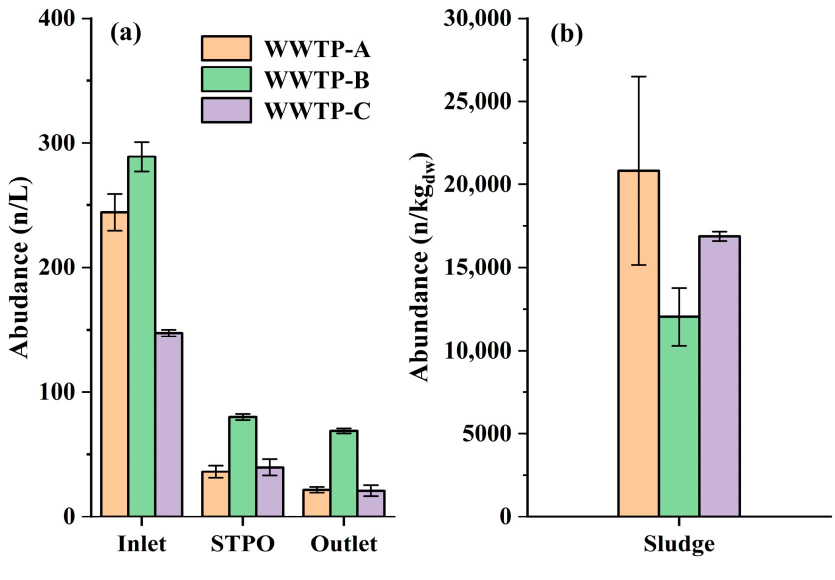

Figure 2. Abundance of MPs in each WWTP (**a**) wastewater samples; (**b**) sludge samples.

The abundance of MPs in the sludge samples appeared to be significantly higher compared to that in the wastewater samples. For sludge samples, due to the substantial number of suspected MPs on each membrane, we employed a random sampling approach in each membrane to select 100 suspected objects for detection and extrapolated the detected proportion to estimate the amount of MPs present on the whole membrane. This number, in relation to the weight of the desiccated sludge specimen, serves as an indicative measure of MP abundance within sludge samples (Figure 2b). In this study, the abundance of MPs in the sludge samples is 20,818.4 ± 5662.0 n/kg_{dw}, 12,024.7 ± 1737.0 n/kg_{dw}, and 16,886.6 ± 274.9 n/kg_{dw}, respectively. Interestingly, our one-way ANOVA showed no statistically significant difference in the abundance of MPs detected in sludge samples obtained from every WWTP (p = 0.107), which seemed to indicate that the abundance of MPs in sludge was relatively stable. However, further research evidence is required to substantiate this claim, as our study employed a random sampling method rather than individually detecting suspected MPs in sludge samples. Our research team is currently developing a support vector machine-based model for suspected object selection, which holds significant implications for enhancing the detection methods of MPs in sludge samples in future studies. Undeniably, sludge in WWTPs serves as an enrichment environment for the accumulation of MPs from wastewater. A case study from one WWTP in the UK showed that the abundance of MPs in sludge ranged from 37,700 to 286,500 n/kg_{dw}, and when the sludge was applied for agricultural purposes, the monthly plastic discharge into the soil corresponded to a volume equivalent to that of two credit cards [29]. However, the study of seven WWTPs in Cadiz revealed that soil amendments derived from sludge contribute to an estimated daily influx of MPs into the environment, ranging from approximately 8.05×10^4 to 1.77×10^9 particles [36]. Undoubtedly, sludge in WWTPs represents a significant source of MPs in the natural environment, particularly within soil. The appropriate management and disposal of the sludge pose a critical challenge that necessitates future attention.

3.2. Size and Shape

In this research, we classified the MPs into three size ranges: 10–50 µm, 50–100 µm, and >100 µm (Table S3), and quantified the relative abundance of MPs within each size category (Figure 3). The predominant size range of MPs detected in both wastewater and sludge samples was observed to be between 10 and 50 µm, constituting over 50% of the total. The prevalence of MPs larger than 100 µm in IN wastewater was particularly evident, with the highest proportion observed in the three WWTPs being only 5.1% (WWTP-C). This is significantly different from a previous study on the size of MPs in drinking water treatment plants [37]. We analyzed that the potential cause lies in the prolonged transportation of urban sewage through the pipe network after it had been treated at the WWTPS, leading to the deposition of certain large-size MPs within the pipes. Furthermore, these larger MPs were also prone to fragmentation into smaller particles due to shear forces exerted by water flow. Similar conclusions had also been drawn from a study investigating the transportation of MPs through pipelines [33].

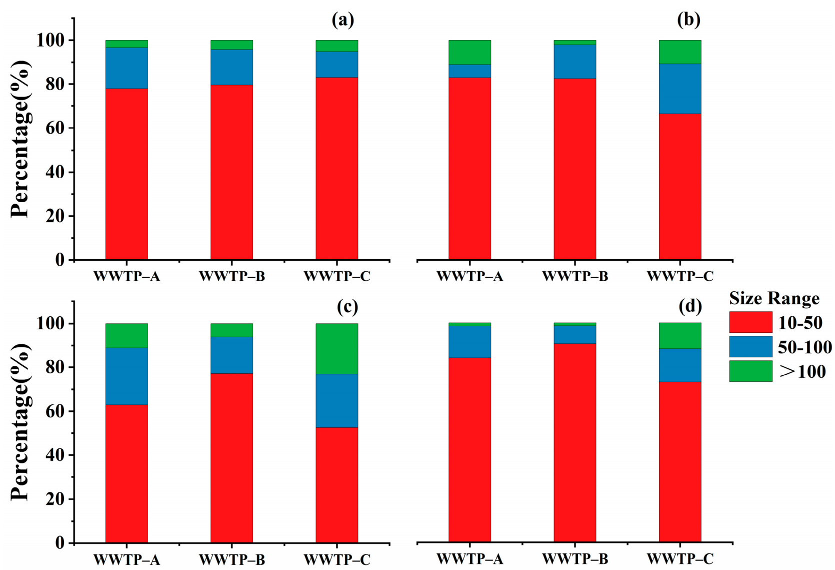

Figure 3. Percentage distribution of MPs' size range in wastewater and sludge samples from each WWTP: (**a**) IN; (**b**) STOP; (**c**) OUT; (**d**) Sludge.

The MPs in other wastewater and sludge samples also predominantly exhibited small dimensions. Some other studies had confirmed that the smallest size ranges generally represent > 70% in observed samples [38,39], the average proportion of MPs from 10 to 50 μm in all wastewater and sludge samples was found to be 76.1%, while the average proportion of MPs smaller than 100 μm reached as high as 92.4% in this study. The presence of smaller-sized MPs poses a challenge for WWTPs, as their removal becomes more difficult [35]. In addition, the average proportion of MPs smaller than 100 μm in the OUT wastewater was found to be 86.5%, while their average proportion in the sludge was determined to be 95.1%. These proportions represent the "sources" of emissions from WWTPs into natural water and soil environments. Furthermore, research has demonstrated that smaller-sized MPs exhibit a higher propensity to infiltrate organisms, thereby inducing visceral impairments and even cellular pathologies [40]. Therefore, it is crucial that future research places greater emphasis on conducting quantitative and sophisticated investigations into the ecotoxicity and environmental impacts associated with smaller-sized MPs.

The MPs detected were categorized into four distinct types: fiber, particle, film, and fragment (Figure 4 and Table S4), and the irregular microspheres detected in this study were incorporated into the statistical distribution of particles. Fiber emerges as the prevailing form, accounting for 81.6%, 86.5%, and 65.8% of the IN wastewater of the three WWTPs, respectively. WWTP-A and WWTP-B primarily cater to the densely populated urban residents of Zhengzhou. Additionally, Zhengzhou serves as a significant inland textile production hub in China, thereby constituting a crucial origin of fibrous MPs. Studies have also demonstrated that domestic laundry and textile manufacturing in industrial facilities can generate substantial quantities of fibers, exerting a notable influence on the morphology of MPs [41–43]. WWTP-C situated in the outskirts, receives sewage that is conveyed through long-distance pipelines. During transportation, MPs undergo mechanical abrasion from external forces and consequently fragmentize, thereby accounting for the relatively higher presence of fragments (19.2%) and small-sized MPs (83.2%). The proportion of fibrous MPs in both STPO and OUT wastewater remained relatively high, with average proportions reaching 78.1% and 83.4%, respectively. This finding further supports previous research indicating the inefficiency of existing WWTPs in removing fibrous MPs [28,44]. As widely acknowledged, a substantial proportion of MPs in inland waters are transported to the ocean via terrigenous sources. Previous research further highlights the ocean's pivotal role as a significant 'sink' for MPs, with fibers remaining the predominant form [45].

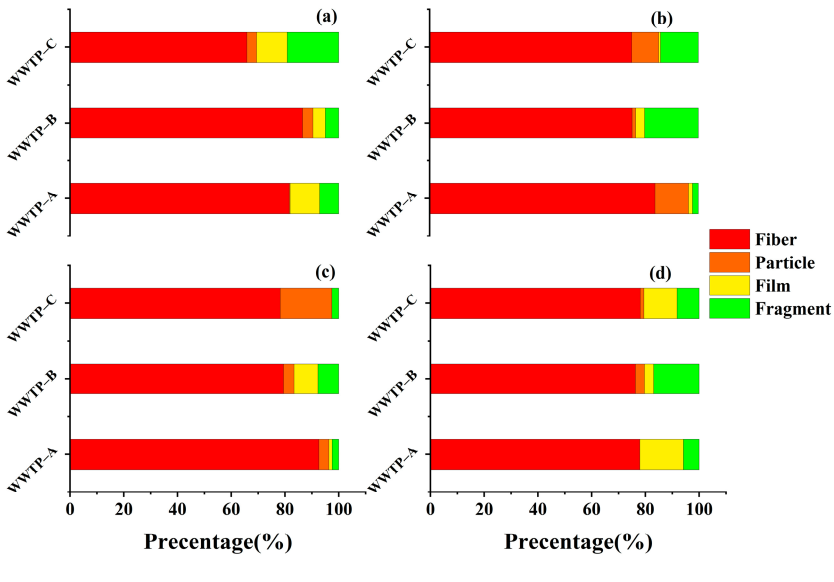

Figure 4. Percentage distribution of the MPs' shapes in wastewater and sludge samples in each WWTP: (**a**) IN; (**b**) STOP; (**c**) OUT; (**d**) sludge.

3.3. Color and Polymer Type

The presence of seven distinct colors of MPs was detected in all samples, namely transparent, gray, blue, yellow, black, red, and green. However, no green MPs were detected in sludge samples (Figure 5 and Table S5). In certain studies, the consideration of color determination was often overlooked in MPs statistics, which posed a limitation as the color of MPs could influence the predation behavior of aquatic organisms [46]. Additionally, the presence of diverse MPs' colors often signifies the incorporation of distinct dye types during plastic production, thereby influencing the efficacy of activated sludge post-precipitation in WWTPs [47]. The transparent MPs constituted the majority, comprising an average proportion of 62.6% in all samples. In general, the proportion of colors could be ranked as follows: transparent > gray > yellow > blue > black > red > green. In the majority of Chinese towns and cities, transparent plastic bags are predominantly used by residents in their daily lives, thereby contributing to the prevalence of transparent MPs. It is noteworthy that the gray color represented the predominant proportion of IN wastewater in WWTP-C, thereby effectively illustrating the significant influence of anthropogenic activities on MPs present in sewage.

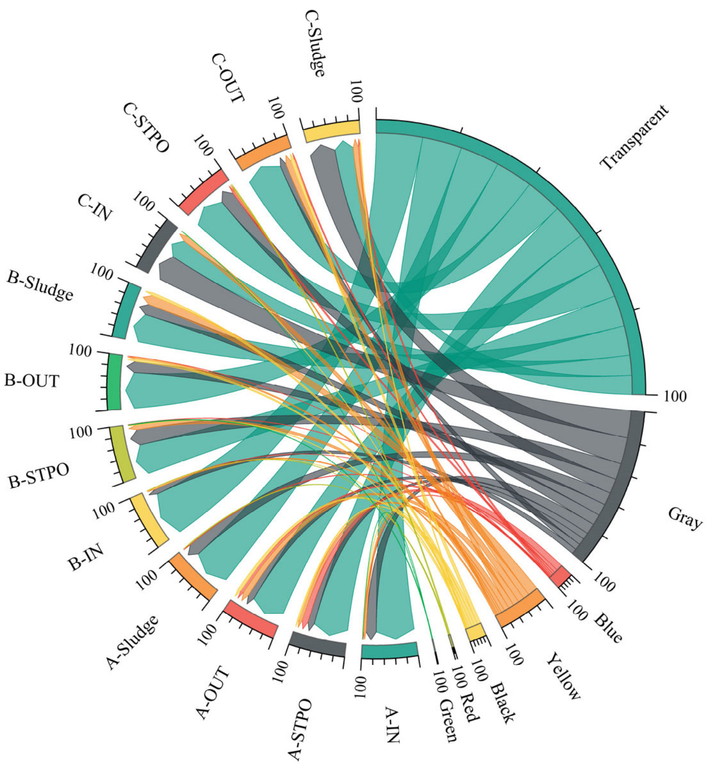

Figure 5. Percentage distribution of the MPs' colors in wastewater and sludge samples in each WWTP.

In addition, no green MPs were observed in sludge samples, and transparent and gray MPs overall accounted for 85.7%. Here, we engage in hypothetical discussions regarding these phenomena: Firstly, this observed phenomenon could be ascribed to the dynamics characteristics of wastewater flow in the operational WWTPs where our sampling was conducted, leading to variations among different sampling locations. Furthermore, the degradation of MPs in sludge is influenced by intricate chemical and biological processes and these alterations are perpetually occurring. Finally, the pretreatment method we used for extracting MPs from sludge samples may possess inherent limitations that could potentially lead to synsemantic extraction. However, it is crucial to acknowledge that our research data solely offers the occurrence of MPs at a stationary time. Therefore, it is imperative to incorporate expanding timelines in future studies to attain more precise analyses.

In total, 10 different polymer types of MPs were identified by a Raman microscopy spectrometer in this study (Figure 6 and Table S6). The average proportions of different polymers in all samples, ranked from highest to lowest, were as follows: polyethylene terephthalate (PET), polypropylene (PP), poly-vinyl chloride (PVC), polyphenylene oxide (PPO), polytetrafluoroethylene (PTFE), polyethylene (PE), polystyrene (PS), polyformaldehyde (POM), polyamide (PA), and polymethyl methacrylate (PMMA). The proportion of PP in the IN wastewater was highest among the three WWTPs, accounting for 76.2%, 53.2%, and 55.2% respectively. A similar trend was observed in the sludge samples, except that WWTP-C exhibited the highest proportion of PET in its sludge, reaching 42.4%. A previous study highlighted that polypropylene (PP) is extensively utilized in China, particularly in the daily catering and medical sectors of the local population [48]. Interestingly, WWTP-A and WWTP-B exhibited the second highest proportion of PPO in IN wastewater, whereas for WWTP-C, it became PET. PET is extensively utilized in the production of various items, including clothing and carpets. Notably, MPs derived from laundry wastewater containing PET represent a significant contributor to the presence of MPs within WWTPs [49], which also indicates that human activities directly affect the composition of MPs in municipal sewage.

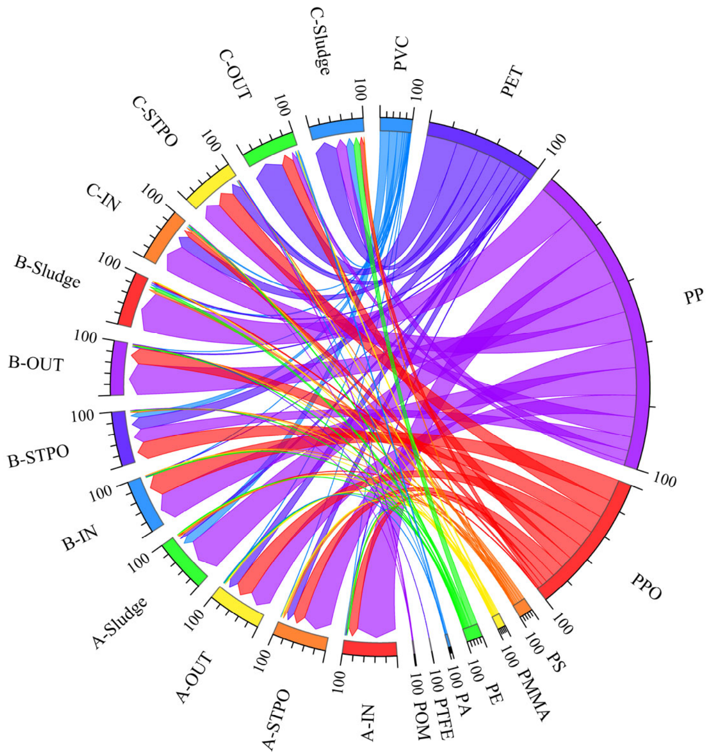

Figure 6. Percentage distribution of the MPs' polymer types in wastewater and sludge samples in each WWTP.

3.4. Removal Efficacy of the MPs

As an emerging contaminant, the water quality standards for effluent from WWTPs do not encompass limits for MPs currently, but relevant research findings have consistently demonstrated the enduring ecological damage caused by it [50]. The detection data in this study can serve as a valuable reference for WWTPs in different regions to investigate the removal efficacy of MPs (Table 1). Here, we calculated the removal efficacy at STPO and OUT, respectively, and the calculation method was as follows:

$$\text{Removal efficacy} = \frac{\text{MPs abundance in IN/STPO} - \text{MPs abundance in STPO/OUT}}{\text{MPs abundance in IN/STPO}} \times 100\% \quad (1)$$

Table 1. MP removal efficacy in each WWTP.

WWTP	Mean Abundance of MPs (n/L)			Removal Efficacy 1 (%)	Removal Efficacy 2 (%)	Removal Efficacy 3 (%)
	IN	STOP	OUT			
A	244.3 ± 14.7	36.3 ± 5.0	21.6 ± 2.4	85.1	91.2	40.5
B	288.8 ± 11.8	80.0 ± 2.4	68.8 ± 2.0	72.3	76.2	14.0
C	147.5 ± 2.6	39.7 ± 6.6	20.8 ± 4.6	73.1	85.9	47.6

Note: removal efficacy 1 represents the removal efficacy of MPs at the STPO; removal efficacy 2 represents the removal efficacy of MPs at the OUT; removal efficacy 3 represents the removal efficacy of MPs in the tertiary treatment process.

The removal efficiencies of MPs in the three WWTPs ranged from 76.2% to 91.2%. WWTP-B showed the highest initial abundance of MPs in influent, yet demonstrated the lowest overall removal efficiency. Furthermore, the removal efficiency 3 of water WWTP-B was merely 14.0%, suggesting that the efficacy of the V-type filter in removing MPs was unsatisfactory. However, further research data are still required to provide a comprehensive explanation. The overall treatment process at WWTP-A is more intricate compared to the other two, resulting in an overall removal efficacy exceeding 90%. The removal of MPs in WWTPs has been demonstrated to primarily rely on the primary and secondary treatment stages in several previous studies [44,51]. The treatment efficiency of various secondary treatment processes on MPs was primarily compared in [52], with the results indicating that SBR exhibited the highest efficacy, followed by AAO, while CAST proved to have the lowest treatment efficiency. The secondary treatment process in this study achieved a removal rate of over 70% for MPs, with WWTP-A demonstrating an impressive proportion of 85.1%. These findings underscore the indispensability of the secondary treatment stage in effectively addressing MPs. Gravity settling and activated sludge adsorption play crucial roles in the removal of MPs during secondary treatment. MPs, regardless of their form, can be effectively adsorbed or captured by sludge flocs and subsequently eliminated through gravity settling [53,54]. The tertiary treatment process in WWTPs bears some resemblance to a part of the treatment process employed in drinking-water treatment plants, primarily relying on various chemical flocculation and filtration. However, the inclusion of sedimentation tanks within drinking-water treatment plants is imperative for the effective elimination of MPs [55]. The absence of sedimentation in the tertiary treatment process of the three WWTPs hinders a clear demonstration of overall MP removal efficiency in this particular stage.

The three WWTPs in this study exhibited an above-average efficacy in MP removal, as evidenced by a comparative analysis with similar studies conducted in other regions (Table 2). A relevant review study suggested that the efficacy of MP removal in WWTPs in China was comparatively lower than that observed in other countries [56], and a study on the Søholt wastewater treatment plant demonstrated a removal efficiency of MPs exceeding 99% [18]. The reason is multifactorial, with a crucial aspect being the lack of standardized protocols for collecting and detecting MPs, which hampers quantitative comparisons across various studies. However, this observation suggests that there is significant potential for improvement in the efficacy of MP removal within certain WWTPs in China. In future research, it is crucial to focus on accurately regulating the operational parameters of existing WWTPs and avoiding extensive construction or renovation of such facilities in order to enhance the efficacy of MP removal.

Table 2. Comparison of MP removal effects between different WWTPs.

Location	Capacity (10^4 m^3/d)	Treatment	Influent (n/L)	Effluent (n/L)	Removal Efficacy (%)	Dominant Polymer Types	References
Hefei/China	10	Primary, secondary (AAO), tertiary	101.9 ± 17.6 (Dry) 108.7 ± 20.1 (Rain)	108.7 ± 20.1 (Dry) 26.3 ± 5.1 (Rain)	87.7 (Dry) 83.5 (Rain)	PA, PF, PE	[57]
Silkeborg/Denmark	105,000 (Person Equivalents)	Primary, secondary (MBR), tertiary	507 ± 70	507 ± 70	99.69	PHDA, PE	[18]
Agadir/Morocco	0.7	secondary (Activated sludge), tertiary	188 ± 29.04	188 ± 29.04	72	PE	[28]
Agadir/Morocco	3.0	Primary, secondary (Filtration-percolation), tertiary	188 ± 29.04	188 ± 29.04	81	PE	[28]
Guiyang/China	1.0	Primary, secondary (SBR/AAO/CAST), tertiary	32.5 ± 1.0	5.0 ± 0.4	84.6	PE, PP	[52]
Changsha/China	0.075	Primary, secondary (AAO), tertiary	70.00 ± 18.67	19.33 ± 1.25	72.4	PE, PS	[32]
Cádiz/Spain (urban)	5.2	Primary, secondary (Activated sludge)	645.03 ± 182.24	16.40 ± 7.85	97	PVC, PA	[58]
Cádiz/Spain (Industrial)	0.008	Primary, secondary (Activated sludge)	1567.49 ± 413.18	1567.49 ± 413.18	91.62	PVC, HDPE, PE	[58]
Wuhan/China	7	Primary, secondary (Activated sludge), tertiary	23.3 ± 2.0	7.9 ± 1.1	66.1	PVC, PE, PP	[59]
Wuhan/China	30	Primary, secondary (Activated sludge), tertiary	80.5 ± 6.3	30.3 ± 3.0	62.7	PVE, PE, PP	[59]

3.5. Limitations

There are still some limitations in this study. Firstly, our study focuses on three WWTPs located in Zhengzhou City, thus making it a geographically limited investigation. The findings of this study can serve as a valuable point of reference for fellow researchers in their respective investigations. Furthermore, it is imperative to acknowledge that our sampling methodology only captures a snapshot in time, thereby limiting our ability to comprehensively investigate the composition and removal efficacy of MPs. The exploration of a certain law necessitates continuous research that extends the research timeline to years in order to obtain more compelling conclusions, which is also the direction we are striving for. Finally, the experimental methodology is established by referencing pertinent prior research findings. We aim to collaborate with fellow researchers in developing a comprehensive and scientifically rigorous set of pretreatment and analysis techniques for MPs, thereby collectively advancing the ongoing research on MP pollutants.

4. Conclusions

In this study, the occurrence and removal efficiency of MPs in three WWTPs in Zhengzhou were investigated. The IN wastewater exhibited an abundance ranging from 147.5 $\pm$ 2.6 to 288.8 $\pm$ 11.8 n/L, and the OUT wastewater showed a range of abundance from 20.8 $\pm$ 4.6 to 68.8 $\pm$ 2.0 n/L. And the abundance of MPs in the sludge samples was 20,818.4 $\pm$ 5662.0 n/kg$_{dw}$, 12,024.7 $\pm$ 1737.0 n/kg$_{dw}$, and 16,886.6 $\pm$ 274.9 n/kg$_{dw}$, respectively. The abundance of MPs detected in wastewater and sludge samples from different WWTPs was calculated by one-way ANOVA, and results showed that there were significant differences among the MPs in wastewater samples between the three WWTPs but not in the sludge samples. The removal efficiencies of MPs in the three WWTPs ranged from 76.2% to 91.2%, and the secondary treatment stage holds paramount importance in the removal of MPs. The predominant constituents of MPs in wastewater and sludge samples were generally observed to be fibers ranging from 10 to 100 μm in size. In total, seven colors were certified in all samples with transparent being predominant. The diversity of polymer types was evident, as a total of ten distinct polymer types were identified across all samples. The proportion of PP in the IN wastewater was highest among the three WWTPs, accounting for 76.2%, 53.2%, and 55.2% respectively. A similar trend was observed in the sludge samples, except that WWTP-C exhibited the highest proportion of PET in its sludge, reaching 42.4%. However, currently, studies on MPs still face a lot of challenges. It is essential to acquire additional basic research data in order to establish a robust foundation for determining limits on MPs in the effluent of WWTPs, thus enabling widespread and systematic analysis of the removal mechanisms of MPs.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/pr12040803/s1, Table S1: Abundance of the MPs in wastewater samples at each WWTP; Table S2: Abundance of the MPs in sludge samples at each WWTP; Table S3: Percentage of the MPs size ranges in wastewater and sludge samples at each WWTP; Table S4: Percentage of the MPs shapes in wastewater and sludge samples at each WWTP; Table S5: Percentage of the MPs colors in wastewater and sludge samples at each WWTP; Table S6: Percentage of the MPs polymer types in wastewater and sludge samples at each WWTP.

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