Evaluation of Microplastic Removal Efficiency at the Wastewater Treatment Plant of a Kraft Paper Factory in Vietnam

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Abstract. Wastewater treatment plants (WWTPs) are one of the critical sinks and sources of microplastics in the environment. Previous studies in Vietnam mainly examined combined treatment plants and centralized plants at industrial zones and lacked data on microplastics in industrial wastewater or sludge. Recycled paper industry wastewater has lots of plastic waste, possibly including microplastics. Therefore, this study aims to evaluate the microplastic pollution load and microplastic removal efficiency of the paper mill wastewater treatment plant. WWTP of the Kraft paper factory A, using primary treatment facilities, level I (Dissolved Air Flotation-DAF), level II (Upflow Anaerobic Sludge Banket-UASB and Conventional Activated Sludge-CAS), and level III (DAF and Fenton), with a capacity of 24,000 m^3 day^{-1} was selected for the survey. The results show that the microplastic load of this factory was 288,000 items day^{-1}, although the removal efficiency was 99.8% and the concentration of microplastics in treated wastewater was 12 items m^{-3}. The primary and secondary treatment processes removed microplastics with the highest efficiency, 75.8–97.9%, especially DAF had a microplastic removal efficiency of $>$95%. Microplastic concentration in the sludge sample was 22,772 items kg^{-1} of dry weight. For morphologies, microplastics in the wastewater and sludge samples existed solely in the form of fragments (55% and 91%) and fibers (44% and 9%), respectively, and blue and white microplastics accounted for 37% and 30% of the total.

Keywords: Microplastics · Kraft paper factory · Activated Sludge · Microplastic Removal Efficiency

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1 Introduction

Wastewater treatment plants (WWTPs) not only act as a barrier but also as a source of microplastics in the water environment in the receiving areas of these wastewater treatment plants [1, 2]. Many studies have been conducted on microplastic contamination in the food web [3], fish [4], water [5], sediments [6], agricultural soils [7], and air [8]. Concerning microplastic pollution in sewage wastewater treatment plants [9], the comprehensive study on microplastic pollution in industrial wastewater and sludge is very few.

In developed countries, microplastics were found in the effluents of WWTPs, although conventional WWTPs with primary and secondary treatment processes can retain 99% of microplastics, and most of them are kept in the pretreatment phases [2]. In particular, the average concentration of 0.05 items L^{-1} of treated wastewater was found from samples collected in 17 different WWTPs in the USA [10]. Ziajahromi et al. also reported similar concentrations of microplastics in treated wastewater samples collected at WWTPs in Australia at 0.2 to 1.5 items L^{-1} [1]. Besides, high concentrations of microplastics in WWTP sludge samples have been revealed in previous studies. Ngo et al. found microplastics in the sludge being 4.20–15.4 $\times$ 10^3 particles kg^{-1} dry sludge [11]. In the study of 28 WWTPs of in China, scientists also reported microplastic concentration in the range of 1.60–56.4 $\times$ 10^3 particles per kilogram of dry sludge [12]. Talvitie et al. reported that the efficiency of microplastic removal after DAF in the WWTPs in Finland could attain up to 95% [2]. Results from a pilot-scale anaerobic treatment system using UASB coupled with anaerobic membrane bioreactor AnMBR presented that 52.6% of the microplastics were removed after UASB [13]. In research conducted by Hu et al., after 16 h of undergoing the degradation process using a thermal Fenton reaction, 95.9% of the microplastic weight was eliminated [14]. In Vietnam, several studies were conducted in the domestic WWTPs [15] and centralized WWTPs of industrial zones [16]. Microplastics were removed from the domestic wastewater after primary and secondary processes of three WWTPs at the rate of 68.8% to 96.7% whereas this rate was 99.9% for a domestic WWTP including tertiary treatment with aerated lagoon followed by a maturation pond [13]. However, in three industrial WWTPs in Danang City, the removal efficiency of microplastics was quite low, varying from 22% to 26% after the primary treatment (grit chamber and primary settling tank) and the secondary treatment [16].

The pulp and paper industry is among the rapidly growing industries [17]. The recycled waste paper used as a raw material for the production of kraft may bring a lot of plastic pieces into the paper mill and then accumulate in sludge. The evaluation of microplastic abundance and its removal is still not well-documented. This paper first evaluates the microplastic removal efficiency at WWTP of a kraft paper factory in Vietnam. Besides, the load of microplastics discharged into the receiving area and their physical characteristics are also identified.

2 Materials and Methods

2.1 Sampling locations

In this study, wastewater and sludge were collected at the WWTP of the Kraft paper factory with a capacity of 24,000 m^3 d^{-1} in June 2020. Wastewater was collected at 8 locations: at the receiving tank (W1), at the equalization tank (W2), after rotating drum (W3), at Dissolved Air Flotation tank (W4), at Upflow anaerobic sludge blanket (UASB) (W5), at the aerobic tank (W6), after the biological settling tank (W7), and after Fenton settling tank (W8), and the sludge were collected at 4 locations at the aerobic tank (S1), the sludge storage tank (S2), the sludge concentrator tank (S3), and the belt sludge press machine (S4). The diagram of the wastewater treatment process at the surveyed WWTP and sampling points was described in Fig. 1.

2.2 Sampling Method

Wastewater samples (from 4 to 400 L, depending on the characteristics of sampling sites) were filtered on-site through a home-made column sieve (a 110-mm diameter PVC tube with a 200 μm-mesh screen) to retain all particles larger than 200 μm in size. Back in the laboratory, for each sample, the particles retained on the screen were carefully rinsed and transferred into a 500 mL glass bottle using tap water (previously filtered through a GF/A paper in the laboratory). Then the bottles were kept in the refrigerator at 4°C until further analysis. Sludge samples (0.5–2 kg) were taken directly using a metal spoon and were stored in a sterile PE Ziploc bag at 4°C.

2.3 Microplastic Identification

The extraction of microplastics from wastewater and sludge samples was carried out based on the treatment protocol proposed by Strady et al. [18], using the same chemical types, dose, and treatment duration. Microplastic observation and measurement of their size, color, and shape were then carried out, using a Leica stereoscope S6D (0.53.15x to 80x magnification) coupled to an HD camera and LAS software. The size of microplastics defined by GESAMP is from 1 to 5,000 μm [19]. In order to identify small-size microplastics, it is required the utilization of modern equipment, such as μFTIR, to define the polymer type of each microplastic. In this research, due to the lack of this equipment, following the recommendation by GESAMP, a minimum length of 300 μm for microplastic fibers and a minimum surface area size of 45,000 μm^2 for microplastic fragments were chosen to identify during the visual observation [19]. Microplastics were identified according to the criteria provided by Noren [20] as follows: (i) the fibers are elongated lines bending in three dimensions with equal thickness throughout the length; (ii) fragments include hard particles with irregular shapes which seem to be fragmented from macroplastics, flat flexible particles having smooth or angular edges, and near-spherical or granular particles which deform readily under pressure and can be partly elastic. Some photos of the microplastic particles observed in this study were shown in Fig. 2.

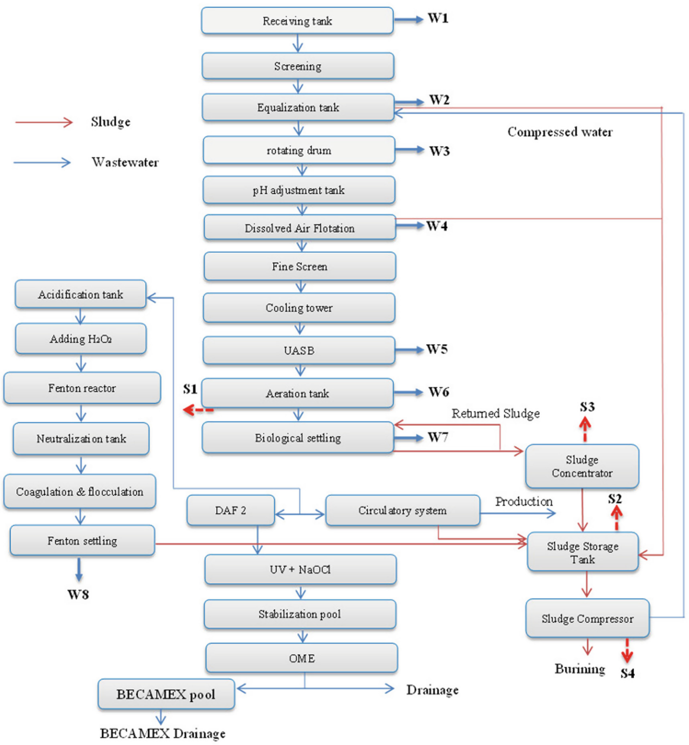

Fig. 1. The diagram of wastewater treatment process at the surveyed WWTP and sampling points

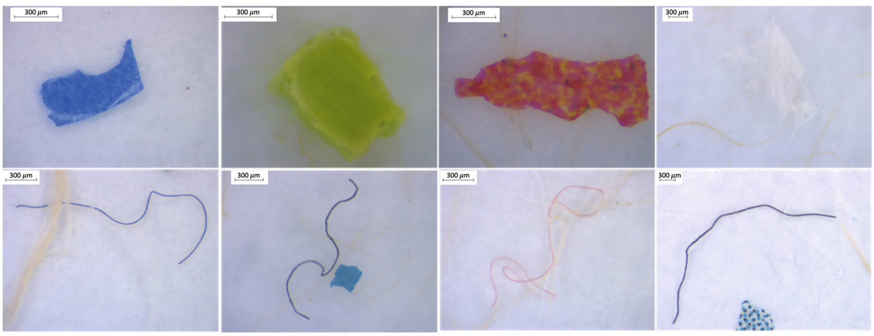

Fig. 2. Stereomicroscopic images of microplastic fragments and fibers observed in wastewaters and sludges

2.4 Data Analysis

The number of microplastic fibers and fragments on the filters was counted and the total, fiber, and fragment concentrations were determined separately for each type of sample. In particular, microplastic concentration was calculated per 1 m^3 of wastewater and per

1 kg of dry sludge, respectively. The XLSTAT® was used for performing microplastics' color partition and illustrating the microplastics' size distribution.

3 Results and Discussions

3.1 Occurrence and Physical Characteristics of Microplastic in Kraft Paper's Mill

Microplastics were found in all wastewater and sludge samples collected at the surveyed WWTP (Fig. 3). The concentration in the inlet W1 was 9,375 items m^{-3}, much smaller than the ones found at five industries studied in Bangladesh, including dyeing, washing, pharmaceuticals, battery, and printing with an average concentration of 2,713,000 items m^{-3} [20]. Microplastic concentration varied at each sampling point during the wastewater treatment process and finally significantly decreased at outlet W8 with 12 items m^{-3}. In sludges, microplastic concentration varied from 7,013 items kg^{-1} of dry weight at the aerobic tank S1 to 22,772 items kg^{-1} of dry weight at the sludge concentrator tank S3, which were 10 times smaller than the results found at five industries in Bangladesh [20].

For morphology, microplastics of different shapes, sizes, and colors were found in the collected samples as shown in Figs. 4 and 5. Two sole shapes of microplastics, i.e., fibers and fragments were also observed in the particles (Fig. 4a) with the predomination of fragments, 55% in wastewaters and 91% in sludges, on average, indicating the different pattern in microplastic shapes compared to the results of Le et al. [15] conducted in domestic wastewater WWTPs where fibers were much more predominant (885 to 99%) than fragments. Seven different colors of microplastics were found in the examined samples i.e., red, blue, grey, white, black, yellow, and green (Fig. 4b). In the inlet wastewater, the dominant color was white (54%) followed by blue (20%) and red and yellow (8% each). Contrary, the dominant color was red (60%) followed by blue (20%) and white (20%) in the treated wastewater. On the other hand, in the sludge samples, the dominant color was observed in white (37%) followed by blue (24%) and yellow (19%). The colors of microplastics indicate associated plastic wrap that might exist in raw materials, recycled paper. Concerning the size of microplastics, at the receiving tank, the microplastic median fiber length is 2,171 μm (Fig. 5a) while the microplastic median fragment area is 192,550 μm^2 (Fig. 5b). These values fluctuate during the treatment process due to the effect of mechanical mechanism such as the flotation and activity of the drum. At the effluent, i.e., the Fenton settling tank, the median fiber length and the median fragment area increase to 2,516 μm and 201,573 μm^2, respectively. The median fiber length of wastewater and sludge of this kraft paper factory was larger than those of 4 domestic WWTPs in the south of Vietnam [15].

3.2 Microplastic Removal Efficiency and Load

The total removal efficiency of microplastics at the surveyed WWTP was 99.9%, higher than the other WWTPs in Vietnam including four domestic WWTPs in Ho Chi Minh City, Binh Duong Province, and Dalat City, and three centralized industrial WWTPs in

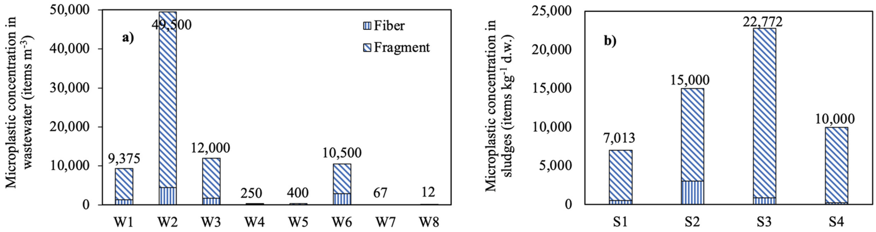

Fig. 3. Microplastic concentration in collected wastewater and sludge samples

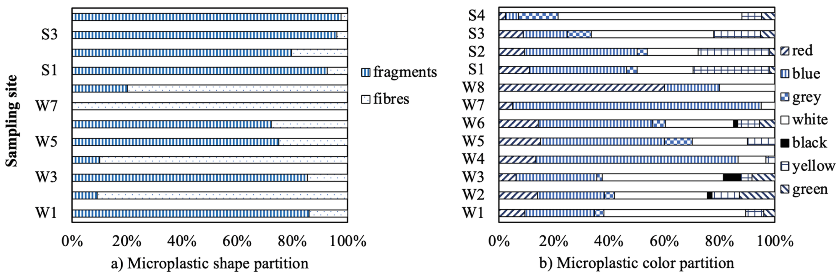

Fig. 4. Microplastic color partition (**a**) and shape partition (**b**) in wastewater and sludge samples

Da Nang City [15, 16]. However, the loading capacity of microplastics discharged to the recipient source, the Ben Trac canal flowing into the Thi Tinh River was also high, with 288,000 items day^{-1}. The primary and secondary treatment units, i.e., the Dissolved Air Flotation W4 and biological settling tank W7, respectively, played important role in removing the microplastics in this kraft paper factory (Table 1), which was contrary to the conclusion of Le et al. [15]. This may come from the difference in characteristics of microplastics observed at the surveyed WWTP where fragments were more predominant.

4 Conclusions

For the first time, microplastic abundance and morphology were evaluated during different treatment phases of a WWTP at a kraft paper factory in Vietnam. Although the technology employed in the studied WWTP is less modern than that in developed countries, the abundance of microplastics in the influent and effluent were 9,357 and 12 items m^{-3}, respectively, leading to the effective removal efficiency being 99,9%. However, the daily loads of microplastic discharged in the receiving system were up to 288,000 items day^{-1}. Fragments were the predominant shape type sharing 55% and 91% of the total microplastics in wastewater and sludge, respectively. This study provides a basis for decision-makers to consider microplastic as potential pollutants to survey in WWTPs from other industries. Besides, the results of this study will serve as a scientific basis for future applied studies on microplastic removal technology in wastewater in Vietnam and provide new scientific knowledge about the source of microplastic pollution in the environment from wastewater.

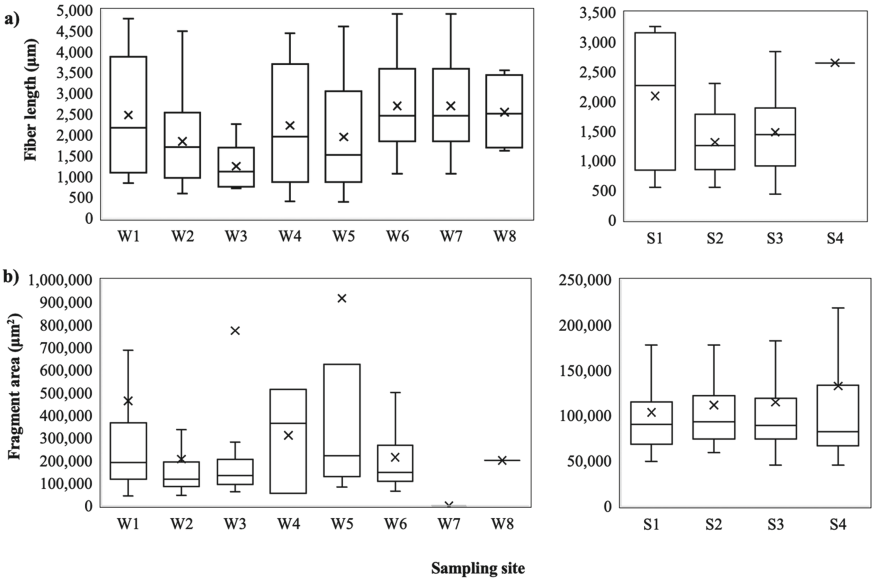

Fig. 5. Boxplots of **a** microplastic fiber length (μm) and **b** microplastic fragment area (μm^2) in wastewater and sludge samples at the surveyed WWTP.

Table 1. Microplastic removal efficiency at treatment steps of the surveyed WWTP

Sampling points	**Microplastic concentration (items m^{-3})**	**Removal Efficiency**
At the equalization tank (W2)	49,500	
After rotating drum (W3)	12,000	76%
At Dissolved Air Flotation tank (W4)	250	98%
At the aerobic tank (W6)	10,500	
After the biological settling tank (W7)	67	99%
After Fenton settling tank (W8)	12	81%

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