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Research Article

Microplastics and solutions to remove microplastics in wastewater from wastewater treatment plants in the Saigon–Dong Nai river basin, Vietnam

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Abstract: The article studies the occurrence of microplastics in the inlet and outlet wastewater streams at wastewater treatment plants in the Saigon-Dong Nai river basin, Vietnam and provide a suitable removal solution. The sampling method is suitable for the actual conditions of Vietnam combined with the application of Fourier-transform infrared spectroscopy to analyze the microplastic composition in the sample. The results show that microplastics exist in many different shapes and colors. Density of microplastics in the inlet stream is from 10.188-15.074 gL^{-1}. Density of microplastics in the outlet stream is from 0.684-2.107 gL^{-1}. In which, filaments with an average length of 524.68 μm and an average radius of 100.4 μm; slender form with an average length of 229.49 μm and an average width of 101.3-120.6 μm; granules with an average radius of 113.81μm. The removal efficiency of microplastics in the wastewater stream at the surveyed wastewater treatment plants ranges from 85.4% to 93.7% through the following main processes: pre-settlement, flotation, moving bed biofilm reactor, sedimentation, filtration. Solutions for the removal of microplastics from wastewater treatment plants in the Saigon - Dong Nai river basin were proposed and discussed.

Keyword: Microplastic; Saigon–Dong Nai river; Wastewater; Wastewater treatment plant.

1. Introduction

Plastic waste has received a lot of attention over the years. The rest of this plastic waste through flows, leaks and discharges wastewater into rivers, seas and lakes, etc [1]. Plastic packaging products for food, beverage and medicinal items are often used only once, which contributes to 61% of the litter on global beaches [2]. Disposable single-use plastic products enter the waste stream shortly after use, contributing to the cumulative accumulation of more than 6.3 billion tons of plastic waste generated worldwide. Only 9% of plastic waste has been recycled globally. Meanwhile, the majority of global plastic waste is either landfilled or ended up polluting the environment (80%). This has resulted in an estimated 4 million to 12 million tons of plastic ending up in the oceans annually [3]. Since 2019, almost the whole world has been, is and will have to struggle with a global pandemic - Covid 19. The World Health Organization has requested a 40% increase in disposable PPE production [4]. If the average global population uses one disposable mask per day, this could lead to monthly global consumption and waste of 129 billion masks and 65 billion gloves [5]. The prolonged

Covid-19 pandemic has caused profound impacts on all aspects of social life, including plastic waste [6]. This pandemic has increased plastic waste [7].

Recent research has shown that in the top ten countries ranked for using plastic in daily life and production, there are eight countries originating from Asia, of which Vietnam ranks 4th [8]. In Vietnam, between 2000 and 13,000 tons of floating plastic debris is collected annually in the main urban canals [9].

Microplastics are persistent, non-biodegradable and cannot be recovered for recycling like large pieces of plastic. Once microplastics are introduced into the environment, they are very difficult to remove. Their classification depends on the intended use of the original plastic. There are many ways to classify. One of them is to classify them using different symbols, including the recycling symbol on the products [10]. Based on size, microplastics are classified as: Macroplastics, Microplastics and Nanoplastics [11].

Wastewater from domestic and industrial activities has been proven to have microplastic pollution. Industrial wastewater and water treatment plants are a major source of microplastic pollution in freshwater ecosystems. Water treatment plants are almost "collection points" of microplastic pollution that are released into the receiving water environment [12-16]. Currently, Vietnam has concluded about the presence of microplastics in the surface water environment and the results discussed about their risk to human health [17].

Wastewater flow under the active control of human is through many different treatment technologies. Following that process, microplastics will be removed. The study by Bayo et al demonstrated that microplastics are removed when the wastewater flows through the primary sedimentation chamber [18]. In addition, microplastics in the waste stream are also removed when going through the stages of flotation [19-20], coagulation, filtration processes such as sand filtration, activated carbon, membrane filtration [21-22], and sludge. activity [23] or membrane technology Membrane Bioreactor (MBR) [24]. To date, relevant evaluation studies have also been published such as the relationship of microplastics and the wastewater treatment system [25] and water ozonation [26]. In general, the removal of microplastics with each treatment technology is still not clear, but the analysis results of microplastics in the inlet and outlet streams of the wastewater treatment system have been published a lot.

Research on microplastic treatment in the world has been carried out by many countries with effective removal: Finland (99.4%), Sweden (99.9%), France (83%-95%), Netherlands (72%), American (99.9%), Germany (97%), Australia (99%), England (98%), Italy (84%), China (97.2%), Russia (95.6%) [27]. The efficiency of removing microplastics from the wastewater stream of some key processes in the wastewater treatment plant: level 1 treatment (58.6%), level 2 treatment (84.1%), level 3 treatment (93.8%) [28-29].

There have been a number of proposed technological schemes to remove microplastics in wastewater such as: diagram of flocculation settling process [30]; diagram of removing microplastics by Anaerobic tank, Anoxic tank, Aerobic tank and Sediment tank [31]; diagram of microplastic collection by pump and filter device [31] or flowcharts for removing microplastics from waste streams with level 1 treatment, level 2 treatment and level 3 treatment processes [24, 32-35]. The objectives of this study are to evaluate the occurrence of microplastics in some wastewater treatment plants in the Saigon - Dong Nai river basin and to propose technical process for removal microplastic.

2. Materials and methods

2.1. Sample collection

Currently, microplastic researchers usually take water samples by using plankton sampling nets to collect surface water samples containing microplastics based on NOAA's Ocean microplastic sampling method [36]. The plankton net sampling method is very suitable for the surface water environment of the ocean, the sampling space is large, the boat connected to the net with the required speed is easy to use. Nonetheless, with the condition of surface water inland rivers of Vietnam, the author takes the sampling method more suitable for the actual conditions as follows:

- At locations with open conditions in the river basin (about 20-50 m from the outlet of wastewater treatment plants in the river basin, depending on actual conditions), surface water samples were collected using a 1×1 m^2 Newton grid, 500 µm meshed meshes 3 meters long. The grids are placed side by side connected by aluminum bars at the top and bottom. The net is connected to the boat by steel wire and a large hook with a flow meter attached, which is used to measure the water velocity at the time of sampling.

- At collection pits or sewers that collect wastewater before entering treatment plants, we use Bucket with wide mouth design to easily scoop surface water (water level thickness is about 30-50 cm from water surface). The water sample consisting of microplastics, impurities and coarse garbage is poured through a sieve of size 0.6-5 mm with a diameter of 300 mm made of 304 stainless steel, passing through the funnel and into the sample containers.

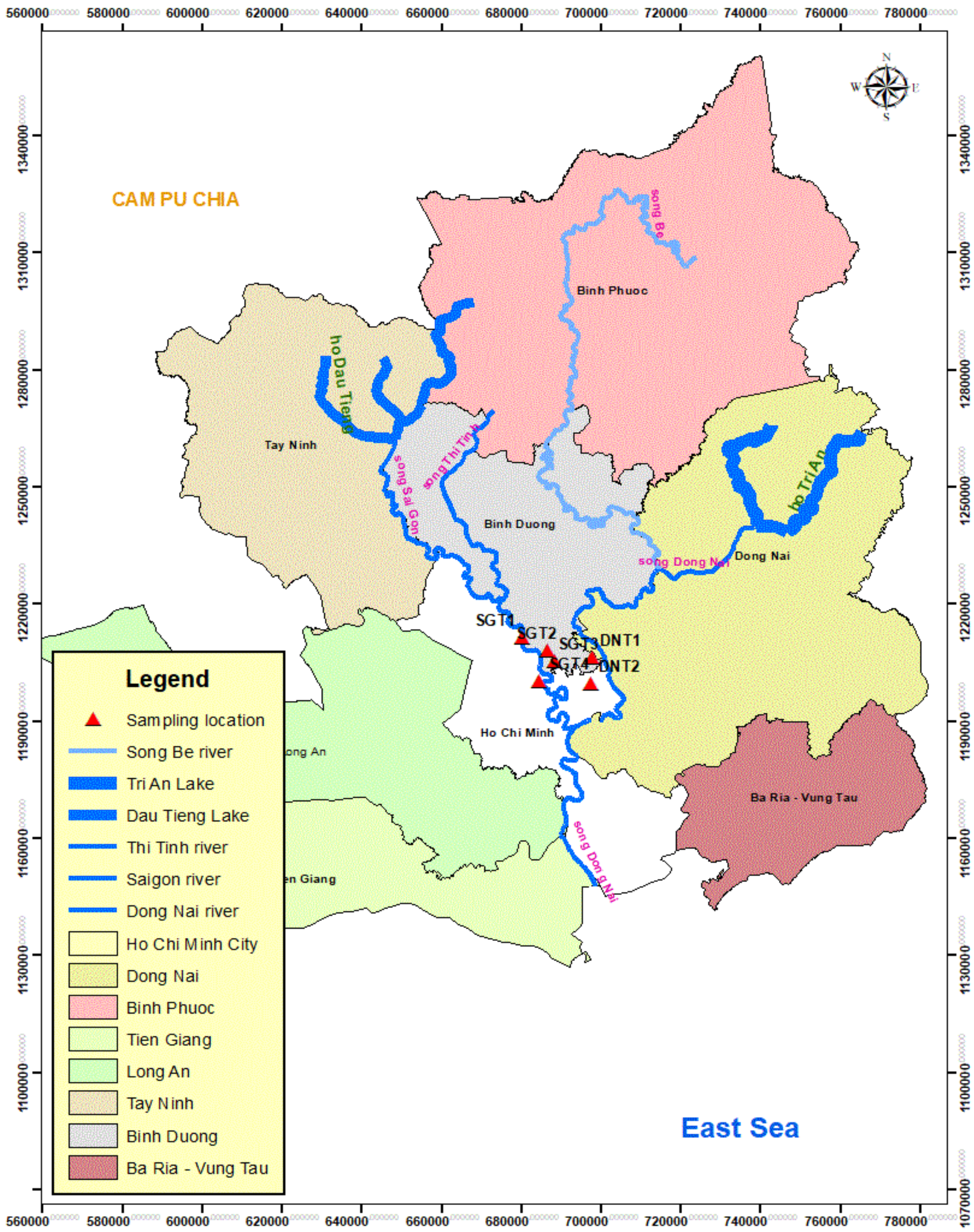

Figure 1. Location map of surveyed wastewater treatment plants.

These surface water sampling locations are flow locations that can carry large amounts of plastic waste from the upper Saigon river and Dong Nai river flowing through densely populated residential areas and industrial zones of Ho Chi Minh City and Binh Duong province. Survey and water sampling were carried out at 6 wastewater treatment plants in the Saigon - Dong Nai river basin, the symbols for the wastewater treatment plants are SGT1, SGT2, SGT3, SGT4, DNT1, DNT2 (Figure 1). The author selected the above 6 wastewater plants for sampling because these plants are located in the area near the end of the Saigon - Dong Nai river basin, at the confluence of 2 rivers. This area is densely populated. These treatment plants mainly treat domestic wastewater for residents of HCM and Binh Duong and have the receiving source in the Saigon-Dong Nai river basin.

Samples will be taken at 02 locations of 6 factories: at the pits and culverts collected before entering the plant and about 20-50m away from the outlet of wastewater treatment plants in the river basin, depending on actual conditions. Sampling time is about 30 minutes per site at low tide. The number of samples to be taken at each location is 2 samples. Sampling frequency is every 6 months in the dry and rainy seasons of the year. In Vietnam, the rainy season is from May to December and the dry season from January to April. Each sample collected at least 2 liters of wastewater containing microplastics. All samples were shipped to the laboratory of Nation Lab Ho Chi Minh City and Phu My Institute of Technology Development for Environment and Water Resources.

2.2. Process of analyzing microplastics in wastewater samples

The collected samples were carefully packed and preserved by the research team in Styrofoam containers before they were transported to the laboratory for analysis (Figures 2a–2b).

Figure 2. (a) Ba Bo canal – Binh Duong, where waste containing microplastics from wastewater treatment plants is discharged; (b) Water samples are ready to be sent to the microplastic analysis laboratory.

Water and sediment samples were coded and stored in Styrofoam and sent to the laboratory for analysis. The process of sample processing and microplastic observation was carried out by the research team according to the steps shown in Figure 3.

Samples after removing coarse impurities larger than 5 mm were dried at 60^{o}C for 24–48 hours. Samples after drying were sieved through 0.3 mm sieve to remove components smaller than 0.3 mm. Samples after 0.3 mm sieving were put into glass tubes (250 mL) and labeled to prepare for decomposition of organic compounds. 20 mL of 30 % H_2O_2 (hydrogen peroxide) solution and 0.05M $FeSO_4$ (Fe II) solution were added to the apparatus (beaker). Let the mixture sit at room temperature for 5 minutes before continuing. The mixture was stirred well, and gently heated on an electric stove 15 minutes (when air bubbles are observed on the surface, remove the beaker from the hotplate and place it in the fume hood until reduced). The mixture was further heated for an additional 30 min. Continue adding another 20 mL of 30% hydrogen peroxide as the reaction changes color from amber to pale yellow.

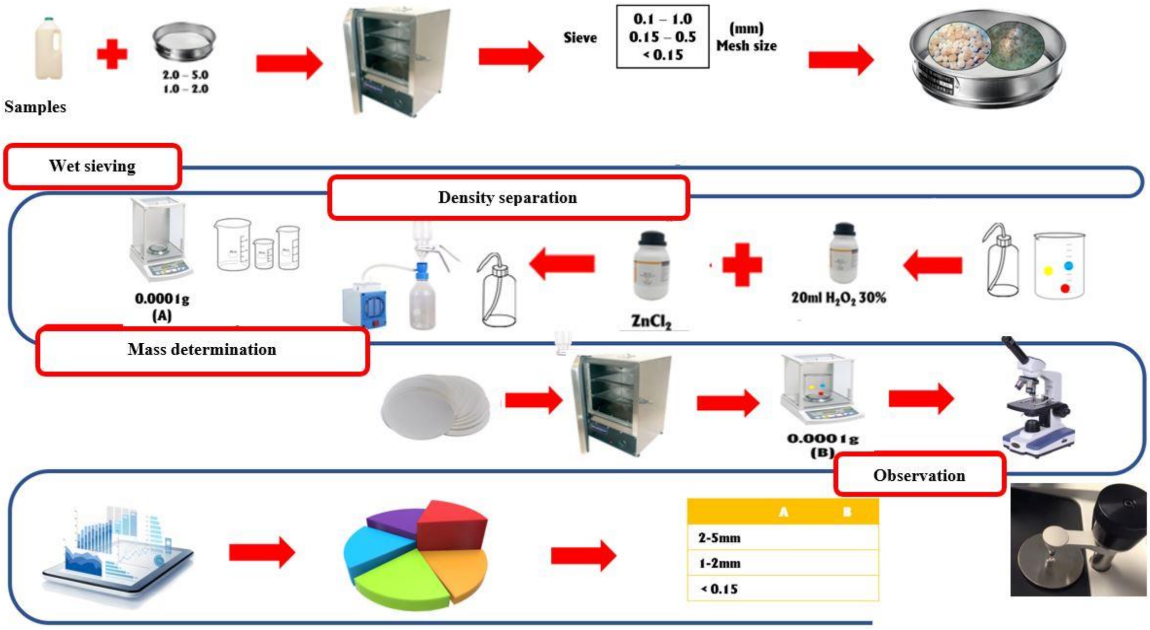

Figure 3. Process of analyzing microplastics in wastewater samples.

To separate minerals and metals: slowly add $ZnCl_2$ solution (d = 1.6 gmL^{-1}) to the sample mixture, stir well, then continue to add $ZnCl_2$ solution in the tube to increase the density of the sample solution. This mixture was put into a centrifuge with a rotational speed of 2500 RCF per min 03 times, for 5 minutes each time to separate microplastics from metals and minerals. Microplastics with a low density will float to the surface of the $ZnCl_2$ solution (minerals and metals with density larger than 1.4 gmL^{-1} will sink at the bottom of the mixture). The supernatant of the mixture was kept for further analysis. The $ZnCl_2$ solution containing microplastics floating above was filtered through the Nalgene vacuum filtration system and used a Milipore reticular filter with 47 mm diameter, 0.45 µm pore size, 3.1×3.1 mm per cell size. Filters were dried and weighed to the nearest 0.1 mg (A_1). The filter is then gently removed and wrapped in aluminum foil bags, dried for about 18–24 hours. The filter after drying is balanced with an accuracy of 0.1 mg (A_2).

Weight of microplastics:

$$A = A_1 - A_2 \tag{1}$$

The identification of microplastics was facilitated using a Fourier transform infrared (FTIR) spectrophotometer.

2.3. Microplastic analysis method by Fourier–transform infrared spectroscopy (FTIR)

FTIR microscopes are generally dedicated to measuring specified samples such as small contaminants on polymer films or microscopic samples transferred to infrared transparent windows. The obtained spectra are compared with internal spectral libraries to find the closest match and determine the chemical composition. A match of 70% or more is considered sufficient for confirmation. The research team applied the FTIR method to determine the microplastic composition in the sample through the spectral peak data obtained when running the sample.

For FTIR analysis, sample vials were washed and poured into a clean, dry, labeled petri dish (separated by size fraction) and placed in a 50°C oven until the petri dish and dry contents. The individual beads were then removed from the petri dish using a microscope (Leica EZ4HD, 8–40× zoom, built–in 3Mpixel camera) and placed on the FTIR (PerkinElmer

Spectrum Two ATR; 450/cm to 4000/cm, 64) scans, resolution 4/cm). The FTIR analysis procedure of the authors is carried out as shown in Figure 4.

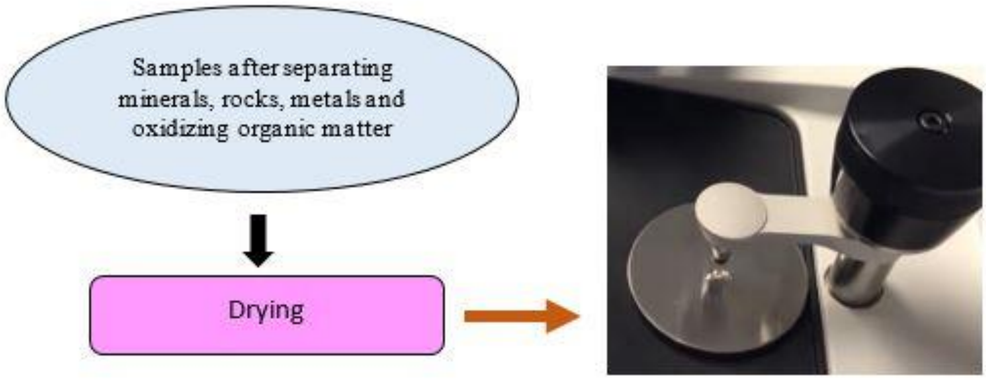

Figure 4. Microplastic analysis procedure by FTIR method.

2.4. Nile Red staining and identification of microplastics by fluorescence microscopy

Fluorescence microscope is a type of biological microscope, which helps to observe the fluorescent light from the specimen after being excited by light from a mercury lamp. When combined with additional equipment, brightfield microscopes can also perform fluorescence imaging. The Nile Red staining method is an alternative to solving the problem of small and transparent microplastics: using the fluorescent dye Nile Red (9–diethylamino5H–benzo[α]phenoxazine–5–one), a strong fluorescence for hydrophobic objects for staining microplastics. The purpose of the Nile Red staining method is to make the resin particles glow more clearly when viewed under a fluorescence microscope. This method helps us to determine the size and density of microplastics in the sample by counting and measuring the size of the luminous particles on the filter paper [35] (Figure 5).

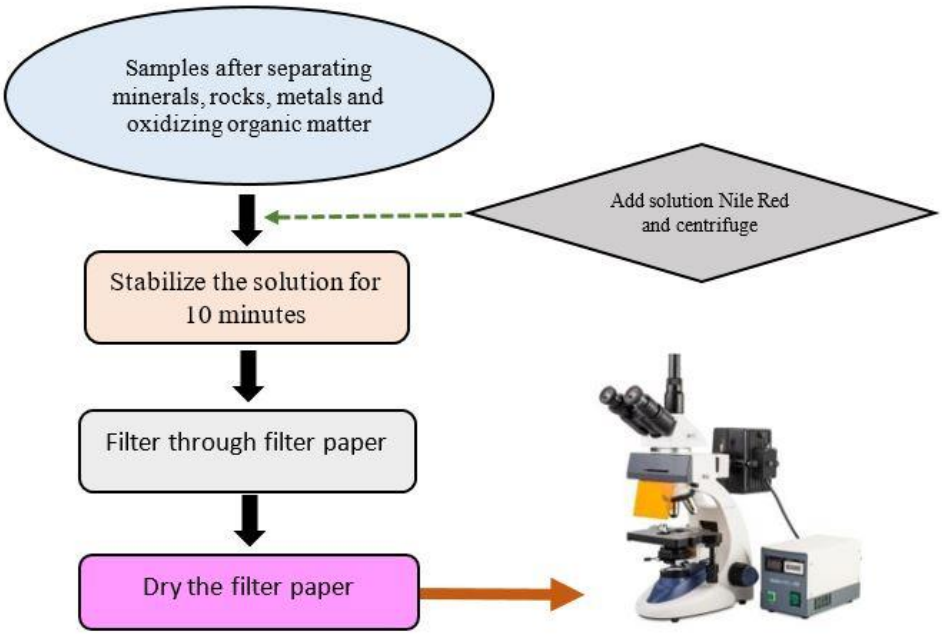

Figure 5. Procedure for analyzing microplastic samples by fluorescence microscopy.

2.5. Method of determining microplastics by stereomicroscopy

The stereo microscope allows for easy 3–D visualization of specimens in their natural state without the need to cut them out. Magnification is usually between 10 and 50 times. The purpose of applying a stereo microscope is that we can observe a 3–D image of the specimen at low magnification. The shape and color of the microplastics were recorded (Figure 6).

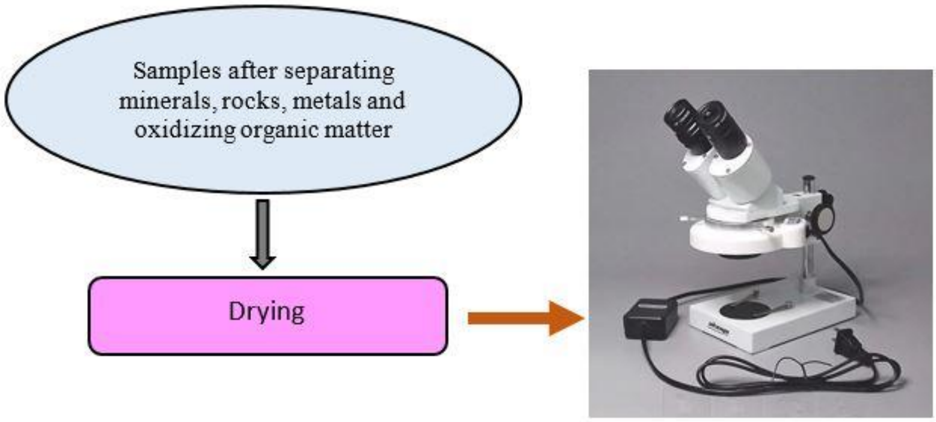

Figure 6. Microplastic analysis process by stereo microscope.

3. Results and discussion

3.1. The results of analysis of microplastics

The results of analysis of microplastics in the inlet and outlet effluents of the domestic wastewater treatment process of some factories in the Saigon–Dong Nai river basin are shown in Table 1.

Table 1. Microplastics in the inlet and outlet waste streams of domestic wastewater treatment in some factories in the Saigon–Dong Nai river basin.

No.	Wastewater plant	Sample symbol	Density of microplastics in the inlet stream (gL^{-1})	Density of microplastics in the outlet stream (gL^{-1})	Removal performance (%)
1	Nam Binh Duong wastewater treatment plant	SGT1	14.432	2.107	85.4
2	Wastewater treatment plant VSIP Industrial Park 1	SGT2	10.188	1.114	89.1
3	Ba Bo Water treatment station	SGT3	12.229	1.516	87.6
4	Tham Luong – Ben Cat wastewater treatment plant	SGT4	15.074	1.749	88.4
5	Di An wastewater treatment plant	DNT1	12.986	1.286	90.1
6	Wastewater treatment plant of Ho Chi Minh City Hi–Tech Park	DNT2	10.851	0.684	93.7
Average performance					**89.1**

Apply a combination of modern microplastic identification methods to be able to more accurately determine microplastic components, more effectively for colorless, transparent, detectable microplastics including microplastics have small and microscopic sizes (Figures 7–10).

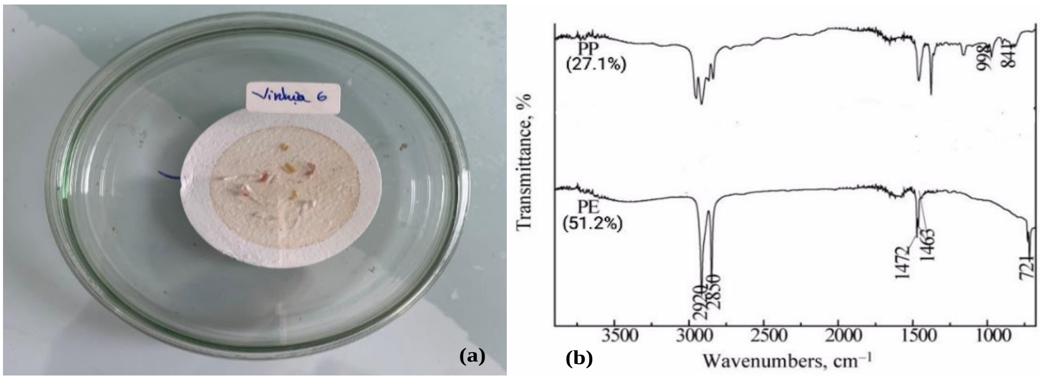

Figure 7. (a) Microplastics were found in the wastewater sample of one wastewater treatment plant (Di An wastewater treatment plant); (b) Spectrum of identification of Polypropylene (PP) and Polyethylene (PE) resins in water by FTIR.

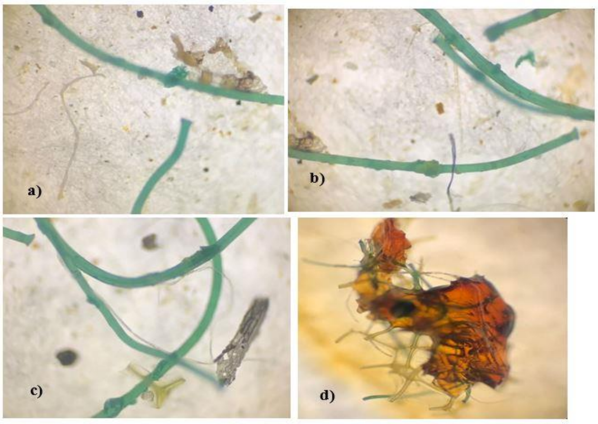

Figure 8. Microscopic filaments under the stereo microscope: (a) Fibrous microplastics are mostly blue > white; (b, c) Microplastics in the form of Fibers of different sizes, blue; d) Microplastic in the form of yellow filaments tangled.

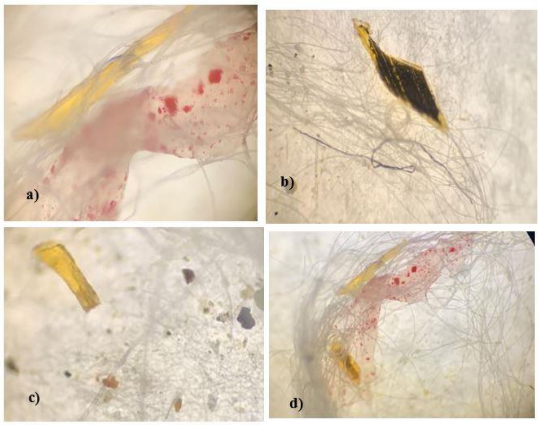

Figure 9. Microplastics in the form of a stereo microscope: (a, b, c) Microplastics of different sizes yellow > green > white; (d) Yellow microplastics surrounded by white fibers.

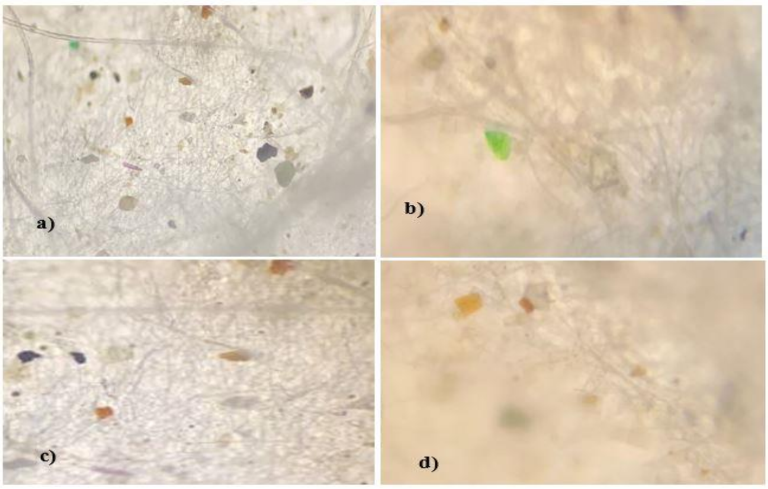

Figure 10. Microplastic granules under a stereo microscope: a) Microplastics with black> blue>white particles; b) Microplastics are green; c) Many microplastics are mixed in white filamentous microplastics; d) Microplastic particles are white.

- The filamentous microplastic has an average length of 524.68 µm and an average radius of 100.4 µm.

- Flake microplastics have an average length of 229.49 µm and an average width of 101.3–120.6 µm.

- Granular microplastics have an average radius of 113.81μm.

Microplastics in the output of domestic wastewater treatment at Sai Gon–Dong Nai river wastewater treatment plants are shown in Table 2.

Table 2. Microplastics in the output stream of domestic wastewater treatment at wastewater treatment plants on Saigon–Dong Nai river.

Number	Wastewater plant	Sample symbol	Fiber form (MPs/Sample)	Flake form (MPs/Sample)	Granular form (MPs/Sample)
1	Nam Binh Duong wastewater treatment plant	SGT1	13	15	12
2	Wastewater treatment plant VSIP Industrial Park 1	SGT2	187	11	15
3	Ba Bo Water treatment station	SGT3	13	55	12
4	Tham Luong – Ben Cat wastewater treatment plant	SGT4	410	13	73
5	Di An wastewater treatment plant	DNT1	10	20	0
6	Wastewater treatment plant of Ho Chi Minh City Hi–Tech Park	DNT2	4	11	2

After the samples were analyzed by the FTIR method, the microplastics in the samples showed that PE accounted for 51.2%, PP accounted for 27.1%, PVC accounted for 13.4% and 8.3% were other plastics. The results of applying analytical methods in Vietnamese conditions show that microplastics have many colors, shapes and very small sizes; microplastics in the form of thin granules, filaments and microplastics ranging in size from 0.1–5 mm.

3.2. Solution for removal microplastics

The technological diagram proposed by the authors to remove microplastics is shown in Figure 11. This diagram is proposed by the author to be applied to domestic wastewater treatment plants to remove microplastics in domestic wastewater; specifically, domestic wastewater of residents on both sides of the Saigon–Dong Nai river basin.

Wastewater containing microplastics from sources is collected into the sump, then transferred to a grease separation tank to remove the amount of grease floating on the water to avoid clogging the pump system and the rear pipeline (this process removes up to 95% microplastics). Wastewater continues to be directed to the conditioning tank to stabilize the flow, concentration and pH balance to suit the operating conditions of microorganisms. After regulating the flow and concentration, the wastewater is transferred to the moving bed biofilm reactor (MBBR) tank to treat organic substances in the water. The outstanding advantage of the MBBR tank is that it saves space and has the ability to handle very well the polluting criteria in wastewater (more than 99% of microplastics are removed). To treat BOD, wastewater is pumped through an aerotank, where an aerator is arranged to create favorable conditions for aerobic microorganisms to grow and increase BOD treatment capacity. Next, the wastewater is taken to a settling tank to settle microbial sludge, where the collected microbial sludge is transferred to a sludge tank and periodically treated by drying or then used as fertilizer. 98% of microplastics are removed as they bind to suspended solids and are separated by sedimentation. Part of the activated sludge will be recycled to the MBBR tank for further treatment.

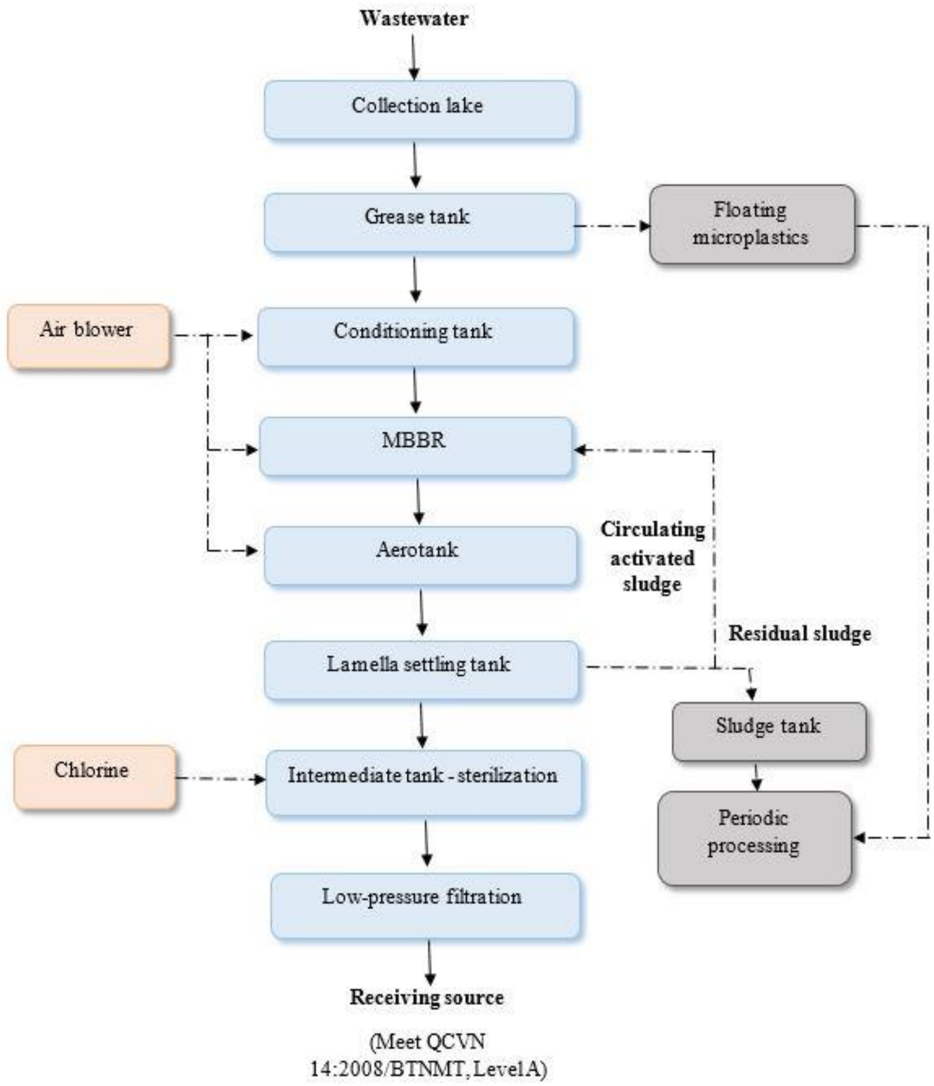

Figure 11. Flowchart of domestic wastewater treatment technology and microplastic removal in wastewater stream.

Wastewater after sludge separation is transferred to a disinfection tank to eliminate all harmful bacteria with chlorine chemicals, then to a pressure filter tank to remove the remaining small residues before being discharged to the receiving source; after being processed to meet QCVN 14:2008/BTNMT column A standards.

Some limitations:

- Requires operator experience;
- Sludge can occur behind the MBBR system according to the biofilm cycle, leading to reduced settling efficiency, reducing microplastic removal efficiency.

4. Conclusion

The average removal efficiency of microplastics in wastewater of treatment plants is from 85.4% to 93.7% through the following processes: pre-settlement, flotation, MBBR, settling and filtration. The development of techniques to remove microplastics from water is necessary to prevent some of the health problems stemming from microplastics. Although the unit works in wastewater treatment technology can partially remove microplastics from the waste stream, but the challenges of technology, minimum cost, efficiency of other components should also be considered in conjunction.

Nonetheless, the limitation of the study is that it is not possible to sample wastewater in each work of the wastewater treatment system. Therefore, the research results only stop at the general assessment of the microplastic removal efficiency of the whole processing when the wastewater flows in and out of the wastewater treatment plant.

Authors contribution. Construction research idea: H.P., H.T.N.H.; Select research methods: H.P., H.T.N.H., N.L.N.T.; T.T.M.H.; Data processing: H.P., H.T.N.H., N.L.N.T.; Sample analysis: H.P., H.T.N.H., N.L.N.T.; Take samples: H.T.N.H., N.L.N.T.; T.T.M.H.; Writing original draft preparation: H.P., H.T.N.H.; N.L.N.T.; T.T.M.H.; Writing review and editing: H.P., H.T.N.H.

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Conflicts of interest. The authors declare that this article was the work of the authors, has not been published elsewhere, has not been copied from previous research; there was no conflict of interest within the author group.

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