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Evaluation of microplastic removal efficiency of wastewater-treatment plants in a developing country, Vietnam

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ABSTRACT

Wastewater-treatment plants (WWTPs) are considered significant point sources of microplastics (MiP) in the receiving waters; MiP release is poorly estimated in developing countries. MiP abundance, recovery efficiency and daily load to receiving waters were explored in treatment stage facilities of four WWTPs in Vietnam, located in Ho Chi Minh City, Thu Dau Mot, and Da Lat. MiP abundance varied from 1860 items m^{-3} to 125,000 items m^{-3} in influents and between 140 items m^{-3} and 813 items m^{-3} in the final effluents. The MiP-removal efficiency was the highest in the Da Lat wastewater-treatment plant (DL WWTP), a tertiary treatment plant using trickling filters followed by aerated lagoon and maturation ponds for BOD and nitrogen removals. The lowest MiP-removal efficiency was observed in Binh Hung wastewater-treatment plant (BH WWTP), a combined sewer system using conventional activated sludge processes as secondary treatment. The total daily MiP load to the receiving environment varied from 1,840,000 items d^{-1} at the DL WWTP to 77,127,000 items d^{-1} at the BH WWTP. Change in MiP abundance, morphology, and removal efficiency were discussed after each stage of the treatment process for the four WWTPs.

1. Introduction

Microplastics (MiP), plastic items in the size range of 1 μm–5000 μm (Kershaw et al., 2019), pose a risk to the health of ecosystems and biota. Although the specific health consequences of this new pollutant are not yet clearly understood, plastic additives, such as bisphenol-A (BPA), and other harmful components, such as trace metals (Foshtomi et al., 2019) and resistant organic matter (Singla et al., 2020; Sørensen et al., 2020), adsorbing to their surface are recognised as potentially harmful to the ecosystem, humans, and other organisms (Di Renzo et al., 2021). Since the first evidence of

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their presence in sediment samples collected from beaches around Plymouth City, England in 2004 (Thompson et al., 2004), microplastics have been found in all environmental compartments, such as the air (Zhang et al., 2020b), the aquatic environments (Xu et al., 2020), soil (Kim et al., 2021), flora (Leifheit et al., 2021) and fauna (Rezania et al., 2018), and sea ice (La Daana et al., 2020). They can even accumulate in the human body, posing health risks (Cox et al., 2019). Their sources and the quantitative analysis of their export from land to sea are key issues that still need to be documented to have a holistic understanding of pollution and to identify the levers to address to decrease pollution. Among the diffuse sources, airborne road and surface runoff play important roles, as do effluents from wastewater-treatment plants (WWTPs), which are major recipients of terrestrial microplastics before they enter natural aquatic systems. The presence of microplastics was found in all tested WWTP effluents (e.g. Hamidian et al., 2021; Sun et al., 2019; Yaseen et al., 2022); despite this, conventional WWTPs with primary and secondary treatment processes can retain 99% of MiPs, mainly during pre-treatment phases (Talvitie et al., 2017). The daily MiP load entering the receiving aquatic environment, mainly rivers, can be important despite the high effective retention rate, as first estimated by Murphy et al. (2016) around 6.5×10^7 microplastics per day on average 261,000 m^3 d^{-1} of treated wastewater. WWTPs function as a barrier and as an entrance route of microplastics to the aquatic environment (Talvitie et al., 2017; Ziajahromi et al., 2017).

To better understand and improve the removal efficiency of WWTPs, efforts have been made to characterise the abundance, morphology and nature of microplastics in WWTPs, and to evaluate the efficiency of microplastics removal from wastewaters based on the removal percentage of MiP at the end of the treatment process (Conley et al., 2019; Hongprasith et al., 2020; Petroody et al., 2020). The influences of MiP was investigated and demonstrated to influence chemical treatment processes by inhibiting the denitrification process (Cluzard et al., 2015), physical treatment processes by increasing the dose of chemicals used for removing suspended solids owing to their large surface area with negative charge (Cluzard et al., 2015), and biological processes by reducing the abundances of functional microorganisms (Zhang and Chen, 2020). Therefore, understanding the performance of MiP retention during the main stages of WWTPs is necessary for clarifying the fate of MiP in WWTPs and considering water reuse for agricultural or domestic purposes, especially in developing countries, where few studies have been conducted so far.

Vietnam, a developing country, is facing severe water pollution mainly caused by untreated wastewaters originating from domestic, industrial, and agricultural activities (Danh, 2021). The total capacity of the current 45 centralised WWTPs cover approximately 12.5% of total domestic wastewater generation rate in Vietnamese urban areas (Danh, 2021). The wastewater-treatment configuration of all WWTPs is used for organic, nutrient and pathogen removal and includes preliminary treatment using bar racks or screens followed by activated sludge processes, such as conventional activated sludge (CAS), sequencing batch reactor (SBR), and oxidation ditch (OD) (Do et al., 2018). These domestic WWTPs were not designed to remove MiP from wastewater, and until now, Vietnamese effluent quality standards have not been based on limited values for MiP concentration. Recently, low MiP-removal efficiencies (22%–25%) have been estimated in three typical Vietnamese centralised (industrial and domestic) WWTPs in Danang City, showing an average loading capacity of MiPs discharged to recipient sources of 3.8×10^7, to 1.5×10^9 particles per day (Van Do et al., 2022), confirming that effluents from domestic Vietnamese WWTPs should be considered as important sources of MiP in the receiving aquatic environment. Therefore, we hypothesised that MiP-removal efficiency might vary according to the treatment process steps used and the type of sewerage system (separated versus combined). This study was designed to evaluate the microplastics retention efficiency of four domestic Vietnamese WWTPs. The objectives were to: (i) evaluate the concentrations and morphologies of MiP in influents, different treatment steps, and WWTP effluents; (ii) investigate the microplastic retention efficiency at each stage of the treatment process; (iii) identify any correlation between physico-chemical parameters and microplastic concentrations; and (iv) estimate the MiP loads released into the aquatic environment from each WWTP.

2. Material and methods

2.1. Sampling sites

Three domestic wastewater-treatment plants (WWTPs) located in Southern Vietnam [Binh Hung (BH), Thuan An (TA), Di An (DA)], and one in Central Vietnam [Da Lat (DL)] were selected to evaluate their MiP removal efficiency (Fig. 1). The treatment capacity, type of sewage system, treatment processes, and effluent quality requirements of each surveyed WWTPs are listed in Table 1. The influents of all WWTPs originate from domestic wastewater generated by residential and service areas. Three WWTPs (i.e. TA, DA, and DL) received wastewater from separated sewage collection systems, while the influent of the BH WWTP came from a combined system collected, in the same sewer line, both domestic wastewater and stormwater. For a combined sewage system, the Vietnamese government requires individual septic tanks for black water pre-treatment of VCA (2014). A conventional activated sludge treatment process is used in the BH WWTP. The WWTPs in the DA and TA use a sequencing batch reactor coupled with a UV channel. Finally, the DL WWTP uses trickling filters followed by aerated lagoons and maturation ponds in their process. At each WWTP, four to five sampling points were selected during the entire treatment process, at key stages, from influent to effluent. The samples were collected in June 2019. The sampling points are listed in Table 1 and shown in Fig. 2.

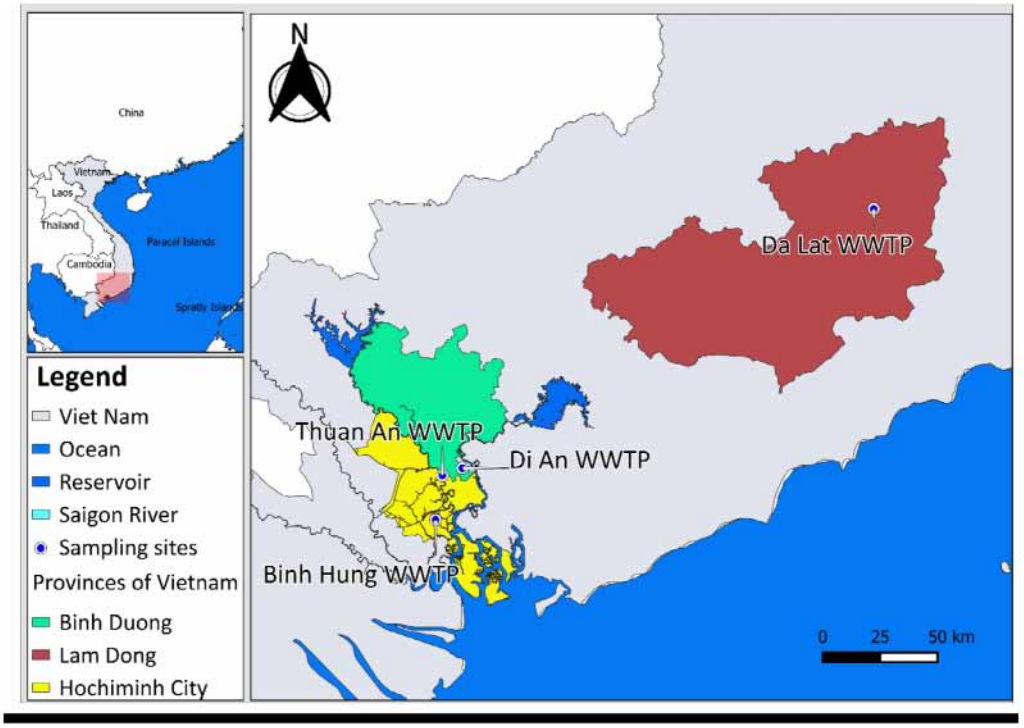

Fig. 1. Locations of four studied WWTPs.

2.2. Sampling method

Discrete grab sampling using a clean stainless-steel bucket of 5 litres was used at all sampling points. Wastewater sample (from 4 to 150 L, depending on the characteristics of sites) (Table 3) was 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. Then, 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). Back in the laboratory, the bottles were kept at refrigerator at 4 °C until further analysis.

2.3. Laboratory analysis

Wastewater samples were analysed using the protocol described by Strady et al. (2021a). Briefly, the main steps were: (i) primary sieving with 1 mm sieve to remove large materials within collected wastewater (ii) sample digestion of the fraction lower than 1 mm with Sodium Dodecyl Sulphate (SDS, Merck®) 1 g L^{-1}, biozym SE (protease and amylase, Spinnrad®) & biozym F (lipase, Spinnrad®) and H_2O_2 (30%, Merck®), (iii) 250 μm sieving of the digested solution to remove the particles smaller than 250 μm in size, then (iv) density separation by doing the overflow, using NaCl solution ($d = 1.18 \pm 0.02$ g mL^{-1}) and finally (v) sample filtration, using the GF/A filters (1.6 μm porosity, Whatman®) and glass filtration unit. The filters were stored separately in a petri dish until observation. To prevent contamination, filtered tap water (through a GF/A filter) was used for the analytical protocols and for cleaning both the working area and glassware. The laboratory staff used cotton laboratory clothes and latex gloves.

2.4. Visual observation and polymer identification

All filters were observed using a Leica Stereoscope S6D (0.5x–80x magnification) coupled to an HD camera. The morphology of microplastics (including size and colour) was measured using integrated LAS software®. According to the criteria suggested by Strady et al. (2021a), microplastic particles are distinguished from other materials by the following rules: (1) microplastic fibres have to be long fibrous material that has a length substantially longer than its width, which is equally thick and has a three-dimensional bending, while (2) microplastic fragments are defined as irregularly shaped hard particles, flat flexible particles with smooth or angular edges, or near spherical or granular particles, which have the

Table 1
Information regarding four surveyed WWTPs (described in Fig. 2).

No	Name of WWTPs	Design capacity (m^3 d^{-1})	Treatment processes	Sewerage system	Location	Effluent quality standard	Sampling points
1	Binh Hung (BH)	141,000	Preliminary: individual septic tank Primary: bar crack, grit chamber, primary clarifier Secondary: aeration tank, secondary clarifier, chlorine contactor	Combined	Binh Chanh District, Ho Chi Minh City	Type B: COD $\leq$ 150 mg L^{-1} TSS $\leq$ 100 mg L^{-1} NH_4^+-N $\leq$ 10 mg L^{-1} TN $\leq$ 40 mg L^{-1}	BH-S1: in front of the bar rack BH-S2: outlet of grit chamber BH-S3: outlet of primary clarifier BH-S4: outlet of secondary clarifier BH-S5: outlet of chlorine contactor as final effluent of the WWTP
2	Thuan An (TA)	17,000	Preliminary: Coarse screen and grit chamber Primary: oil and grease trap Secondary: Sequencing batch reactor, UV channel	Separated	Thuan An City, Binh Duong province	Type A: COD $\leq$ 75 mg L^{-1} TSS $\leq$ 50 mg L^{-1} NH_4^+-N $\leq$ 5 mg L^{-1} TN $<$ 20 mg L^{-1}	TA-S1: in front of coarse screen TA-S2: outlet of grit chamber TA-S3: outlet of oil and grease trap TA-S4: outlet of UV channel as final effluent
3	Di An (DA)	20,000	Preliminary: coarse screen, grit chamber Primary: oil and grease trap Secondary: sequencing batch reactor, UV channel	Separated	Di An town, Binh Duong province		DA-S1: in front of the coarse screen DA-S2: outlet of grit chamber DA-S3: outlet of oil and grease trap DA-S4: outlet of UV channel as final effluent of WWTP
4	Da Lat (DL)	12,000	Preliminary: bar crack, grit chamber, Primary: Imhoff settling tank Secondary: trickling filter, secondary clarifier Tertiary: aerated lagoon, maturation pond, chlorine contactor	Separated	Da Lat City, Lam Dong province		DL-S1: in front of bar rack DL-S2: outlet of grit chamber DL-S3: outlet of Imhoff settling tank DL-S4: outlet of secondary clarifier DL-S5: outlet of chlorine contactor as final effluent

(*) QCVN 40:2011/BTNMT: Effluent quality standards issued by Ministry of Natural Resource and Environment – Vietnam.

appearance of being broken down from larger litter and deformed readily under pressure or partly elastic. For the internal structure, all microplastic particles should have an absence of internal repetitive identical structures (organic structure) and should exhibit a clear and homogeneous colour throughout. The observation size range was [300–5000 μm] for the fibres, [45,000–25,000,000 μm^2] for the fragments.

The chemical composition of 203 particles (including 124 fibres and 79 fragments) was identified using an FTIR-ATR iS50 Thermo Fisher Scientific® (CASE laboratory, HCMC, Vietnam). The tested particles represented only a small percentage of the total number of observed particles. Therefore, particles were chosen within regard to their representativeness of the sample selection as a whole, to our uncertainty regarding their visual observation and classification as a plastic polymer, and to their possibility of being easily picked up and analysed by FTIR-ATR. Observed particles were all plastic polymers, particularly polyethylene PE (36%), polypropylene PP (22%), polyethersulfone PES (21%), acrylic (8%) and other types for the rest.

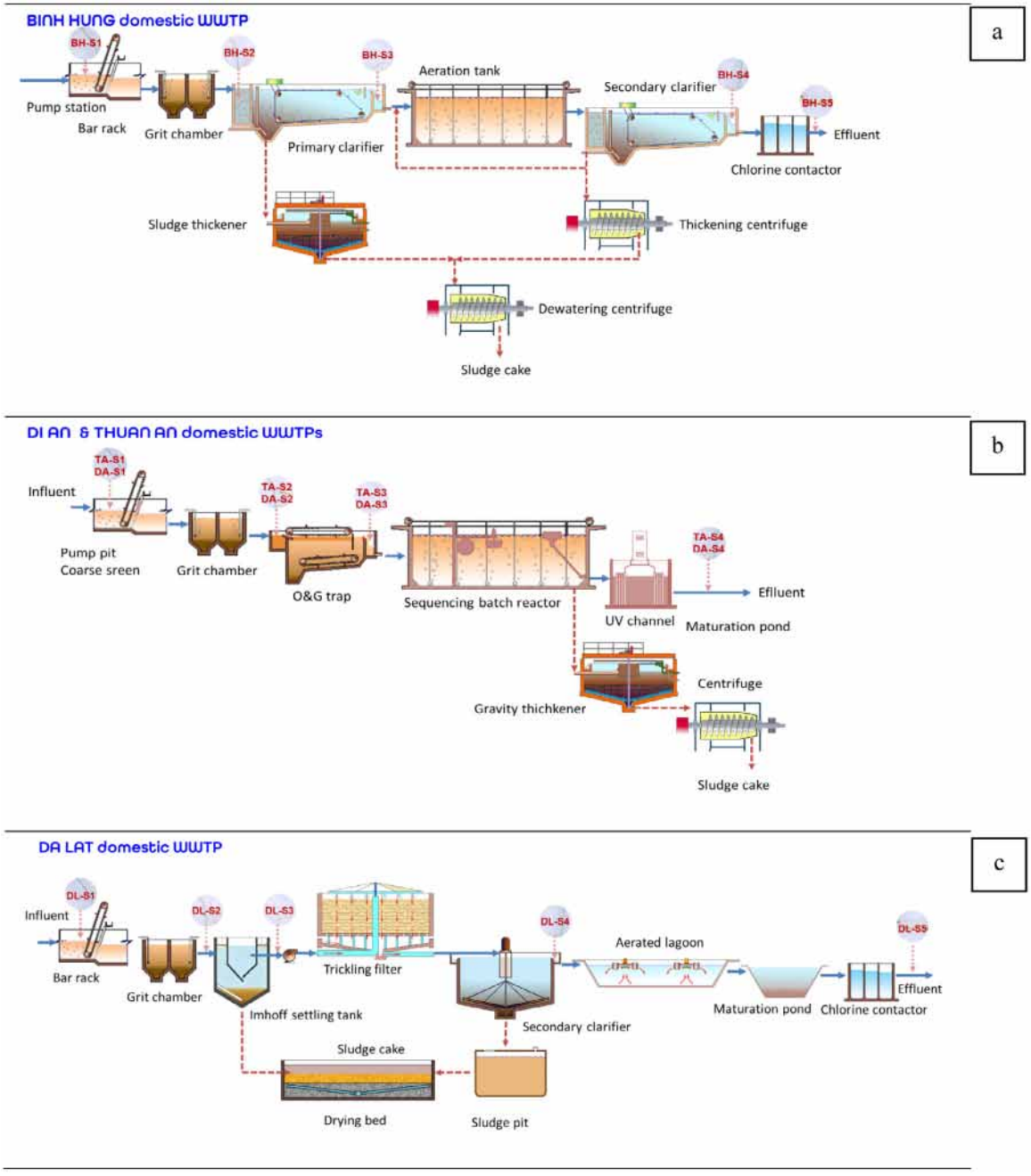

Fig. 2. Diagram illustrating treatment stages and sampling points of four studied WWTPs.

2.5. Chemical analysis

Total Suspended Solids (TSS), soluble Chemical Oxygen Demand (sCOD), and total Chemical Oxygen Demand (tCOD) were measured in the laboratory on five samples from the BH WWTP, four samples from the TA WWTP, four samples from the DA WWTP and five samples from the DL WWTP. TSS was measured by employing the 2450D method and samples were filtered through pre-weighed Whatman GF/F filter membranes 0.7 μm and oven-dried at 103–105 °C for 24 h, and re-weighed to measure the dry TSS weight. tCOD and sCOD were analysed using the closed reflux method (Standard Method 5220C APHA AWWA, 2005), with prior filtration through 0.7 μm membrane (GF/F, Whatman™, UK) for sCOD parameter. From these parameters, particulate COD (pCOD) was calculated using the following equation: pCOD = tCOD − sCOD.

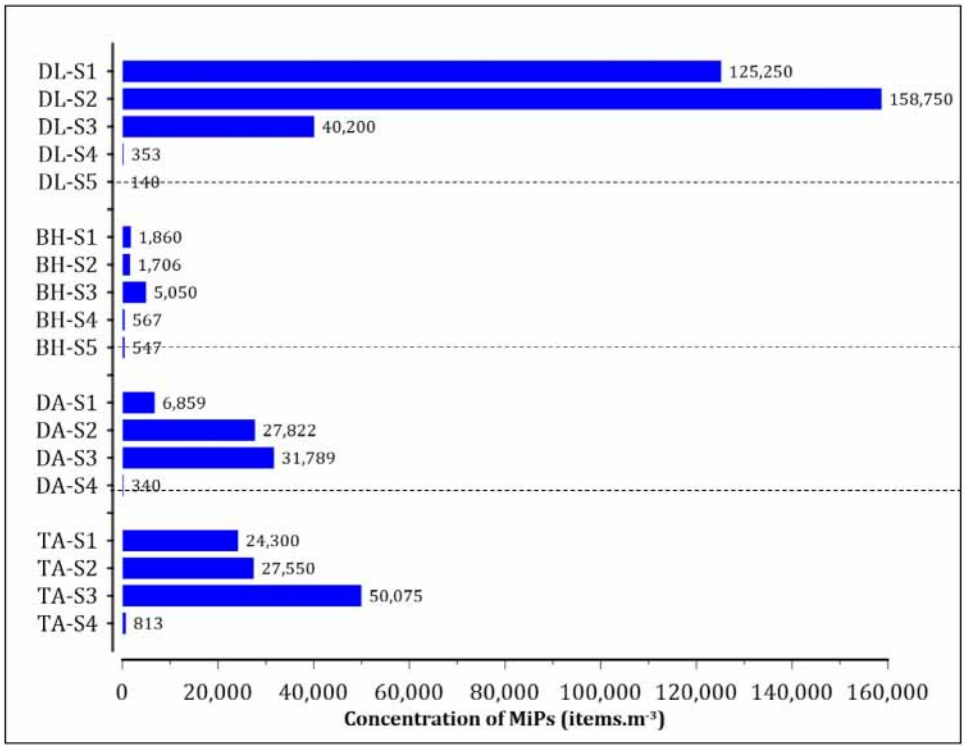

Fig. 3. Concentrations of MiP in each step of the treatment process studied in four domestic WWTPs.

2.6. Data analysis

The number of microplastic fibres and fragments on the filters were counted, and the total microplastic, fibre, and fragment concentrations were expressed by item m^{-3}. XLSTAT® software was used to perform MiP colour partitioning and illustrate the size distribution of MiP. The raw data were synthesised by a Python Script using Anaconda and Jupyter Notebook program packages.

2.7. Calculation of microplastic and TSS removal efficiency

The MiP and TSS removal efficiencies (RE) were calculated using the following equations:

$$RE(\%) = \frac{(C_i - C_e) \times 100}{C_i}$$

where C_i is the concentration of MiP or TSS at the outlet of the oil and grease trap at the TA and DA WWTPs, an Imhoff settling tank at the DL WWTPs, and a primary clarifier at the BH WWTPs.

C_e is the concentration of MiP or TSS in the final effluent at the TA, DA and DL WWTPs and the outlet of the secondary clarifier DL and BH WWTPs.

3. Results and discussion

3.1. Concentration, morphology, and colour of MiP in wastewaters entering the preliminary steps and in effluents

3.1.1. Mip concentrations

MiP concentrations measured in wastewaters before the bar rack (BH-S1, DL-S1) and before the coarse screen (TA-S1, DA-S1) varied from 1860 items m^{-3} at the BH WWTP to 125,000 items m^{-3} at the DL WWTP (Fig. 3). Concentrations measured in BH, DA, and TA WWTPs were in the same concentration range as observed in WWTP from France (2.9×10^5; size range: 100–5000 μm) (Michielssen et al., 2016), Thailand (12.2×10^3; size range: 330–475 μm) (Hongprasith et al., 2020), UK (15.7×10^3; size range: 2800 μm) (Murphy et al., 2016), while concentration measured in DL WWTP was in the same range as that measured in the USA (133×10^3; size range: > 100 μm) (Michielssen et al., 2016) (Table 2). The difference in MiP concentrations measured in Vietnamese wastewater entering the preliminary steps may be attributed

Table 2
Concentrations of MiP reported in WWTPs worldwide.

Country		Capacity (m^3 d^{-1})	MiP concentration		MiP observation size range	Average retention efficiency	Treatment processes	Reference
			Influent (items m^{-3})	Effluent (items m^{-3})				
Thailand		200,000	12.2×10^3	2×10^3	Fibre: 330–475 μm	83.6%	Primary + secondary	Hongprasith et al. (2020)
South Korea		26,545	4.2×10^6	33×10^3		89.9%	Primary + Secondary + Coagulation + Ozone	Hidayaturrahman and Lee (2019)
USA		2,500,000	133×10^3	5.9×10^3	Microbeads: > 100 μm	98.5%	Preliminary + Primary + Secondary + Discfilter	Michielssen et al. (2016)
France		240,000	2.9×10^5	32×10^3	Fibres:100–5000 μm	94%	Preliminary + Primary + Secondary	Dris et al. (2015)
UK		260,954	15.7×10^3	0.25×10^3	Fibres: 2800 μm	73.5%	Primary + Secondary + anaero-bic/anoxic/aerobic	Murphy et al. (2016)
Australia		13,000	N/A	0.28×10^3	Fibres: 25–500 μm	> 90%	Primary + Secondary + Tertiary	Ziajahromi et al. (2017)
Vietnam	**TA**	**17,000**	**24.3×10^3**	**0.81×10^3**	**Fibres: 300–5000 μm Fragments: 45,000–25,000,000 μm^2**	**96.7%**	**Preliminary + Primary + Secondary**	**This study**
	DA	**20,000**	**6.9×10^3**	**0.34×10^3**		**95%**		
	BH	**141,000**	**1.9×10^3**	**0.58×10^3**		**68.8%**		
	DL	**7500**	**125.25×10^3**	**0.14×10^3**		**99.9%**	**Preliminary + Primary + Secondary + Tertiary**	

N/A: not available

to the type of sewer system and the respective domestic water demands and, inputs from the residents (Long et al., 2019; Tang et al., 2020). The lowest concentrations were measured at site BH-S1 of the BH WWTP, in waters coming from a combined sewer system during the rainy season. The receiving waters at BH-S1 are thus a mix of wastewater and surface urban runoff waters, which have been demonstrated as a major pathway for microplastics (Werbowski et al., 2021; Wang et al., 2022), especially when the microplastic content in the dust road is elevated (Yukioka et al., 2020). However, the water dilution induced by the large volume of water entering the system owing to the rainy season coupled with the presence of individual septic tanks may have enhanced microparticle dilution and settling and reduced their inputs (TSS inputs measured at 31 mg L^{-1} at BH-S1) to the influent and to BH-S1. In contrast, the high MiP abundance measured at DL-S1 may be due to the lower water demand of residents, lower infiltration rate, or different inputs from the residents, and its cause should be investigated in a future study.

In the final effluents (TA-S4, DA-S4, BH-S5, DL-S5), MiP concentrations ranged from 140 items m^{-3} at the DL WWTP to 813 items m^{-3} at the BH WWTP, and concentrations measured in the final effluents were systematically lower than those in the respective wastewaters entering the preliminary steps (TA-S1, DA-S1, BH-S1, DL-S1) (Fig. 3). The concentrations measured in Vietnamese effluents were in the same range as those measured in other countries using biological treatment processes, such as activated sludge or trickling, for the secondary treatment level (Table 2). Notably, with similar analytical and observation protocols, concentrations measured in Vietnamese WWTP's effluents are significantly higher than those measured in their receiving waters such as the Dong Nai River (6.5 ± 3.7 items m^{-3}) or Tri An reservoir (1.5 ± 0.6 items m^{-3}) (Strady et al., 2021a). This demonstrates that WWTPs are a source of MiP to their receiving environments and specifically to the Dong Nai river and Tri An reservoir.

3.1.2. Mip morphology

The potential effects of the WWTPs treatment steps on the morphology (shape, colour, and size) were addressed. Fragments and fibres were the two shapes observed in the WWTPs, while pellets were not observed. Fibres dominated, with a minimum of 76%, and the percentage of fragments ranged from non-detection (DL-S3) to 24% (DA-S4). The proportion of fibres in all effluents was lower than that in the untreated wastewater, and thus an opposite observation was made for the fragment. We suggest that fibres could be more easily trapped in bioflocs or biofilms during the biological treatment step and that their settling could increase in the secondary clarifier; Fibre and fragment removals in the biological treatment step were fibre and fragment removals of the biological treatment step were respectively 96% and 93%. More specifically, a significant change in the proportions of fragments and fibres was observed in the final effluent samples (TA-S4 and DA-S4).

Regarding fibres, the evolution of their size along the WWTP steps were studied by calculating the fibre's length median, minimum, maximum, first, and third quartiles at each studied treatment step of each WWTP, followed by the Kruskall–Wallis test followed by the Conover–Iman test (Table 3 and Fig. 4). At the DL WWTP, no variation in median fibre length was observed between the final effluent and wastewater entering the preliminary steps (DL-S5 = DL-S1 = a, Fig. 4)

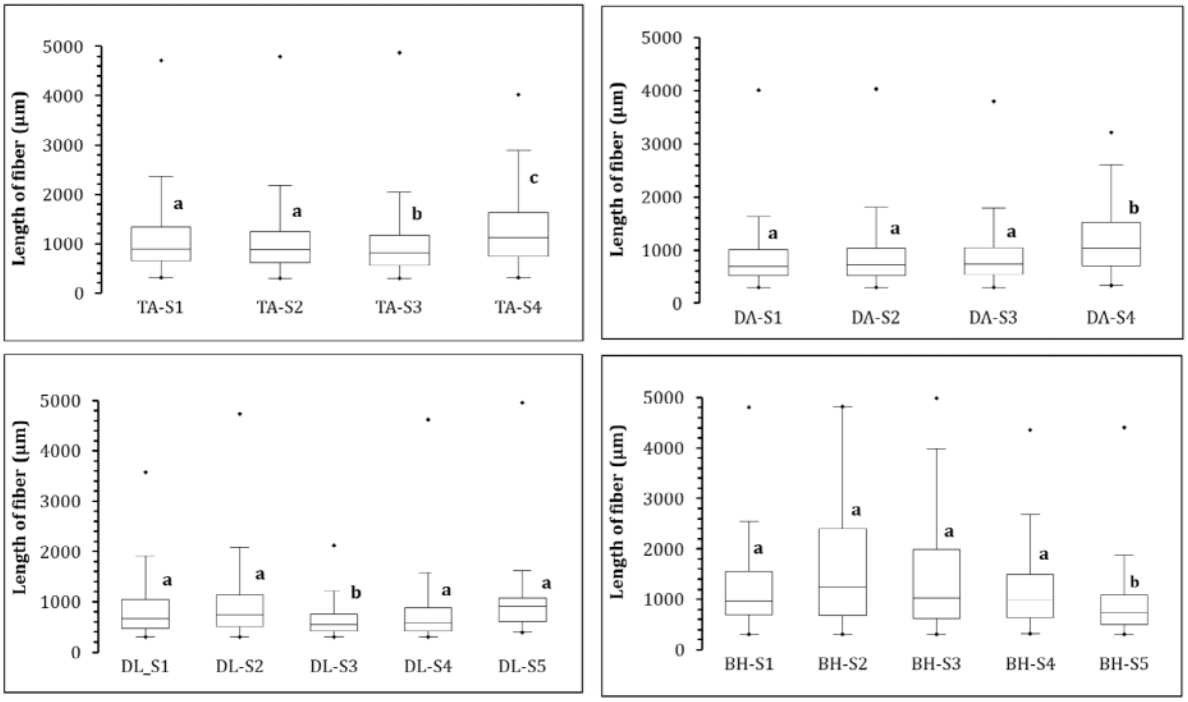

Fig. 4. Colour and morphology repartition of fibre and fragments in all wastewater samples. Letters refer to Kruskall–Wallis test followed by the Conover–Iman test, with a significant level at p=0.05.

Table 3
Colour and morphology repartition of fibres and fragments in all wastewater samples.

ID	Volume	MiP repartition		Fibre length			Fragment area			Fibre colour repartition							Fragment colour repartition							Retention
name	sampled L	Fibre %	Fragment %	Median µm	Min µm	Max µm	Median µm²	Min µm²	Max µm²	Blue %	Red %	Black %	Green %	Grey %	Yellow %	Transparent %	Blue %	Red %	Black %	Green %	Grey %	Yellow %	Transparent %	efficiency %
TA-S1	50	94	6	893	309	4723	82 461	45 639	874 984	49	34	0	5	6	2	3	45	11	12	8	1	1	22	96,7
TA-S2	40	98	2	879	302	4795	144 804	48 140	640 253	68	22	nd	3	2	2	3	30	30	5	15	nd	nd	20	
TA-S3	40	98	2	805	300	4875	95 732	45 534	367 570	67	22	nd	4	3	2	2	40	27	nd	29	nd	nd	4	
TA-S4	150	88	12	1129	311	4020	114 627	53 981	658 304	54	37	nd	nd	nd	1	7	43	36	nd	14	nd	nd	7	
DA-S1	38	91	9	688	302	4009	156 210	50 725	910 327	40	41	1	2	nd	nd	16	39	22	30	9	nd	nd	nd	95,0
DA-S2	18	97	3	727	301	4041	167 252	45 116	710 449	72	22	nd	1	3	3	0	29	50	nd	14	nd	7	nd	
DA-S3	19	96	4	734	300	3805	80 647	45 350	2 755 163	72	16	1	3	5	2	1	32	27	nd	18	nd	9	14	
DA-S4	150	76	24	1026	329	3213	124 962	54 261	437 740	36	54	nd	5	nd	3	3	58	17	8	8	nd	8	nd	
BH-S1	50	94	6	958	315	4807	119 765	45 012	738 553	72	21	nd	5	2	nd	nd	17	50	nd	17	nd	17	nd	68,8
BH-S2	51	92	8	1248	306	4822	388 886	186 313	793 129	56	28	nd	8	1	1	6	71	14	nd	nd	nd	nd	14	
BH-S3	20	98	2	1034	313	4989	984 012	123 725	1 844 298	57	26	nd	14	2	1	nd	nd	50	nd	nd	nd	nd	50	
BH-S4	150	98	2	990	321	4354	193 003	69 095	316 911	69	20	nd	5	1	4	1	50	50	nd	nd	nd	nd	nd	
BH-S5	150	94	6	728	308	4400	145 230	50 450	378 534	82	12	1	2	nd	2	nd	20	60	nd	20	nd	nd	nd	
DL-S1	4	98	2	668	302	3582	186 458	45 415	1 277 486	56	26	3	2	8	2	3	25	8	8	8	8	17	25	99,9
DL-S2	4	100	0	746	301	4734	117 397	90 839	143 955	70	19	nd	2	4	3	1	nd	100	nd	nd	nd	nd	nd	
DL-S3	5	100	0	549	300	2121	nd	nd	nd	65	26	3	1	2	1	1	nd	nd	nd	nd	nd	nd	nd	
DL-S4	150	94	6	673	303	4533	75 311	62 870	1 003 935	64	26	nd	nd	4	4	2	100	nd	nd	nd	nd	nd	nd	
DL-S5	150	91	9	910	395	4956	169 748	125 882	213 614	62	24	nd	nd	5	nd	10	100	nd	nd	nd	nd	nd	nd	

despite a slight decrease in the median size during the primary treatment (outlet of the Imhoff settling tank) (DL-S3 = b, Fig. 4). This decrease in the median fibre length could be due to the absorption of large fibre in the sludge settling during the primary sedimentation step. At the TA, DA and BH WWTPs, the median fibre lengths determined at the final effluents (TA-S4, DA-S4, and BH-S5) were significantly different from the median fibre length determined in wastewater entering the preliminary steps at the TA-S1, DA-S1, and BH-S1 sites ($p < 0.05$). More precisely, at DA and TA, characterised by separated sewer system, the median fibre size in surface wastewaters is increasing significantly ($p < 0.05$, Fig. 4) while at BH, the combined sewer system, the median fibre size in surface wastewaters is decreasing significantly ($p < 0.05$, Fig. 4). It is also important to note that at BH-S1, the median fibre length was longer (e.g. 958 µm, Table 3) than at TA-S1 (e.g. 893 µm, Table 3) and DA-S1(e.g. 688 µm, Table 3). The variation in fibre size distribution along each WWTP clearly show that the applied treatment processes in the TA, DA, and BH WWTPs influence the MiP size distribution in surface wastewaters; in some cases, smaller fibre sizes preferentially settle or are trapped, while in others, larger fibre sizes are preferentially settling or trapped. Settling experiments in columns under laboratory conditions have shown that fibre settling orientation, fibre diameter, and fibre length influence the settling velocity of fibres (Nguyen et al., 2022); hence,

Table 4
MiP-removal efficiency (RE^{MiP}), TSS concentrations and TSS-removal efficiency (RE^{TSS}) after secondary and tertiary treatment at four surveyed WWTPs.

WWTPs	Treatment levels	Sample ID	MiP concentration (items m^{-3})	RE_{MiP} (%)	TSS (mg L^{-1})	RE_{TSS} (%)
TA	Secondary treatment	Outlet of an oil and grease trap (TA-S3)	50,075	98	66	83
		The final effluent (TA-S4)	813		11	
DA		Outlet of an oil and grease trap (DA-S3)	31,789	99	178	92
		The final effluent (DA-S4)	340		15	
BH		Outlet of a primary clarifier (BH-S3)	5050	89	146	95
		Outlet of a secondary clarifier (BH-S4)	567		8	
DL		Outlet Imhoff settling tank (DL-S3)	40,200	99	325	95
		Outlet of a secondary clarifier (DL-S4)	353		15	
DL	Tertiary treatment	Outlet of a secondary clarifier (DL-S4)	353	60	15	27
		The final effluent (DL-S5)	140		11	

it is very difficult to point out the processes responsible for the change in fibre size distribution surface in wastewaters along the WWTP's steps.

3.1.3. Mip colours

Seven colours were observed in all the domestic wastewater samples (Table 3). Blue and red were predominant colours of fibre-shaped MiP in the all samples, accounting for 36%–82% and 12%–54%, respectively, and blue and transparent colour dominated in fragment-shaped MiP in TA and DL WWTP. The blue/red fibre-shaped MiP may be generated from synthetic cloth rags or textile washing, whereas transparent fragment-shaped MiP may result from fragmented macroplastic waste in surface run-off reaching the sewer system. Indeed, a large quantity of synthetic cloth rags and macroplastic bags have been found in sewage and city inner canals in Vietnam (Lahens et al., 2018). The variety of MiP colours in domestic wastewater from different cities may be attributed to a wide range of sources (Gallagher et al., 2016). Lahens et al. (2018) stated that the predominance of blue colour was detected in water samples from urban canals the HCMC and Saigon River, which receives the effluents of BH, DA, and TA WWTPs.

3.2. Mip-removal efficiency

MiP-removal efficiency of the whole treatment processes was evaluated at 97% for TA WWTP, 95% for DA WWTP, 69% for BH WWTP, and 99% for DL WWTP (Table 3). The MiP-removal efficiencies of the three treatment levels of the four WWTPs were also evaluated in this study using the formula defined in Section 2.7. The preliminary and primary treatment levels include the bar rack, screen, grit chamber, oil, and grease separator, whereas the primary clarifier was used in the surveyed WWTPs to remove coarse and suspended solids. Secondary treatment levels, such as the activated sludge process (in BH, TA, and DA WWTPs), and trickling filters are used for BOD and nitrogen removal, and the tertiary treatment level at DL-WWTP only includes aerated lagoons followed by maturation ponds to remove further nutrients and suspended solids.

3.2.1. Effects of preliminary/primary treatment levels on MiP-removal

The concentrations of MiP measured at the outlet of the grit chamber (DA-S2, TA-S2, and DL-S2) and outlet of the oil and grease traps (DA-S3 and TA-S3) were higher than those in the untreated wastewater samples (DL-S1, TA-S1, and DA-S1), indicating that MiP retention by the preliminary/primary treatment level was not efficient (Fig. 3). In the scientific literature, both the effectiveness (Dris et al., 2015; Michielssen et al., 2016; Murphy et al., 2016; Ziajahromi et al., 2021) and the lack of effectiveness (Hongprasith et al., 2020) of the primary treatment for MiP retention have been observed. Zhang et al. (2020a) deepened the effect treatment parameter setting and demonstrated the velocity and travelling distance of the water flow influence MiP removal. MiP would be eliminated if its settling time was smaller than the distance divided by velocity and if its density was higher than 2.6 g cm^{-3} (Zhang et al., 2020a). At DL, at the outlet of the Imhoff settling tank (DL-S3), the MiP concentration decreased significantly after this process, along with decrease in the median fibre length. We can thus hypothesise that microplastics, including most large fibres, settled vertically in the tank during this step.

3.2.2. Effects of secondary treatment level on MiP-removal

Recently, secondary treatment processes have been identified because of high effectiveness in MiP-removal (Lares et al., 2018; Lee and Kim, 2018; Talvitie et al., 2017). A similar pattern was observed in this study during the conventional activated sludge step at the BH WWTP, the sequencing batch reactor step at the TA and DA WWTPs, and the trickling filter step at the DL WWTP in comparison to other treatment levels.

At TA and DA WWTPs, from the outlet of oil and grease trap (TA-S3, DA-S3) to the final effluents (TA-S4, DA-S4), MiP-removal efficiency were of 98% and 99% respectively (Table 4). At BH WWTP, from the outlet of primary clarifier

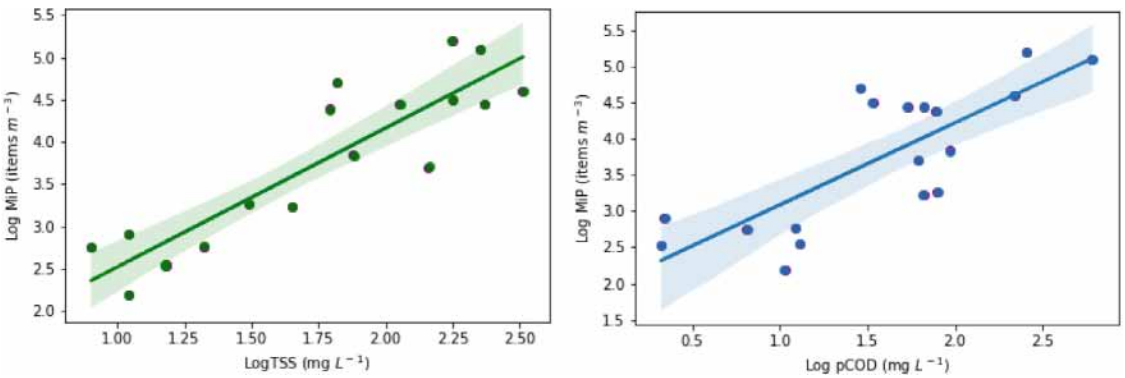

Fig. 5. Relationship between TSS and MiP (left side) and pCOD and MiP (right side) of all samples measured at four studied WWTPs.

(BH-S3) to the outlet of secondary clarifier (BH-S4), MiP-removal efficiency was 89% (Table 4). At DL WWTP, MiP-removal efficiency from the outlet Imhoff settling tank (DL-S3) to the outlet of the secondary clarifier (DL-S4) was estimated to be 99%. After the secondary clarifier (BH and DA WWTP), biological flocs and biofilms may entrap MiP debris and settle together (Magni et al., 2019; Murphy et al., 2016; Peng et al., 2014).

In fact, the observed significant positive correlation between TSS and MiP ($r > 0.9$ and $p < 0.0001$; Fig. 5) illustrates the high performance of TSS removal (e.g. range of 83%–95%, Table 4) after secondary treatment processes in the four WWTPs which may flocculate MiP and remarkably reduce MiP loads entering the receiving waters. Schmiedgruber et al. (2019) stated that approximately 99% of fibre MiP was entrapped into mixed liquor in the activated sludge process; they observed that, as well as Frehland et al. (2020), fibre MiP concentrations increased with TSS depth profile of the settled sludge layer. In our study, MiP concentrations in the mixed liquor of SBR of TA and DA, and in CAS of BH were 7.1×10^5, 6.6×10^5, and 1.0×10^5 items m^{-3}, respectively; MiP concentrations in the outlet of the secondary clarifier at TA, DA and BH WWTPs were 831, 340, and 567 items^{-3}, respectively. Hence, we suggest that the MiP captured in the bioflocs and biofilms was transferred to the sludge cake, as already by Ngo et al. (2019).

3.2.3. Effects of tertiary treatment on MiP-removal

Among the four WWTPs, only the DL-WWTP used an aerated lagoon followed by a maturation pond as the tertiary treatment stage (Fig. 2c) to improve the removal of nitrogen and TSS. From the outlet of secondary clarifier (DL-S4) to final effluent (DL-S5), MiP-removal efficiency was measured at approximately 60% (Table 4). This removal efficiency is lower than that calculated using rapid sand filtration (RSF), dissolved air flotation (DAF), or membrane separation (MBR) (removal efficiency above 95%) (Talvitie et al., 2017). The efficiency of these technologies could probably be because membrane filtration acts as a selective barrier, allowing only certain substances, such as molecules, ions, or small particles, to pass through. With RSF technology, MiP was apprehended among sand grains or attached to the surface of the sand grains. Meanwhile, the DAF process also significantly decreases MiP, which has a lower density than water, because it is designed to eliminate low-density matter (Talvitie et al., 2017). Thus, compared with MBR, RSF, and DAF technologies, it can be seen that maturation pond does not significantly reduce the quantity of MiP in the final effluents at the DL WWTP.

In summary, WWTPs with high BOD or TSS removal by activated sludge or trickling filters obtained high MiP-removal efficiencies, claiming that WWTPs with high-performance biological treatment facilities can significantly minimise MiP pollution in the receiving waters. The differences in removal efficiency between the surveyed WWTPs may be due to the performance of operating primary and secondary clarifiers and, control of the settleability of biofilm and bioflocs. Proper operating measures to prevent resuspension of the entrapped MiP from sludge/scum, such as (i) frequently withdrawal of primary sludge, biosludge, or scum from the clarifiers, (ii) uniform effluent collection by weir adjustment, and (iii) proper control of sludge depth in the clarifiers may enhance MiP-removal efficiency.

3.3. Daily MiP loads discharged into the receiving water

The MiP discharge loads from domestic wastewater treatment plants in receiving water bodies were calculated based on the MiP concentrations measured at each final effluent and the respective daily volume of effluents released by each WWTP. We estimated a total daily MiP discharge load of 13,826,000 items d^{-1} at TA, 6,800,000 items d^{-1} at DA, 1,840,000 items d^{-1} at DL and 77,127,000 items d^{-1} at BH domestic WWTPs. These daily MiP loads are in the same range as worldwide WWTP as reviewed by Liu et al. (2021) (e.g. 500,000–14,000,000,000 items d^{-1}) and more specifically to Australian domestic WWTPs (e.g. 10,000,000 items d^{-1} (Ziajahromi et al., 2017)), Scottish domestic WWTPs (e.g. 65,000,000 items d^{-1} (Murphy et al., 2016)), or Israeli domestic WWTP (e.g. 220,000,000 items d^{-1} Ben-David et al., 2021). In the four studied WWTPs, the highest daily MiP load was calculated at the BH domestic WWTP (e.g. 77,127,000 items d^{-1}). The BH domestic WWTP receives domestic wastewaters from more than 425,000 inhabitants, compared to

100,000 inhabitants for TA, 40,000 inhabitants for DA and 53,000 inhabitants for DL. Furthermore, BH is the only one with combined sewer system, and thus receives atmospheric deposition coupled with surface road dust that is drained in the sewer system while raining or road cleaning with water. In HCMC, deposition flux of microplastics varied between 71 and 917 items m^{-2} d^{-1} (Strady et al., 2021b), which was among the highest deposition fluxes observed worldwide while the road dust, measured not in HCMC but in Da Nang City, the third biggest city in Vietnam, presented MiP concentrations of 19.7 $\pm$13.7 items m^{-2}. Consistent with the finding of Uddin et al. (2020) on the role of population density and consumption ratios of personal care and cosmetic products by inhabitants on final MiP loads, we suggest that microplastics originating from atmospheric deposition and road dust contributes to its high MiP daily load to the receiving waters, the BH influent.

Finally, this study demonstrated that MiP could not be adequately removed by the physical and biological processes used in the four domestic WWTPs studied. Therefore, MiP should be placed in the priority pollutant list to be monitored in WWTPs to control the release and adapt the setting parameters of each step to improve the MiP-removal efficiency.

4. Conclusions

For the first time, microplastic abundance and morphology were evaluated during different treatment phases in Vietnamese WWTPs. Although the technology employed in the studied WWTPs is less modern than that in developed countries, the abundance in the effluents, removal efficiency coefficient, and daily loads of MiP discharged in the receiving system are in the same range as other WWTPs worldwide. The observed MiP size range considered omitted smaller plastics, which are the most abundant. Therefore, this study is a first step in evaluating the state of MiP-removal efficiency and loads in the aquatic environment in Vietnam. It allows WWTP managers and technicians to determine technological efficiency, especially at each step and to optimise it. This study also provides a basis for decision-makers to consider microplastic as potential pollutants to survey in WWTPs. In the future, scientific investigations should address broader observation size ranges (e.g. lower size), to detect representative polymers in samples (more than 50% compared to a few now) and also address MiP fate during flocculation, settling, and resuspension during specific treatment steps.

CRediT authorship contribution statement

Thi-Minh-Tam Le: Validation, Formal analysis, Investigation, Data curation, Writing – original draft, Writing – review & editing, Visualization. **Tran-Nguyen-Sang Truong:** Methodology, Resources, Formal analysis, Investigation. **Phuoc-Dan Nguyen:** Investigation, Resources, Writing – review & editing. **Quang-Do-Thanh Le:** Investigation, Resources. **Quoc-Viet Tran:** Investigation, Resources. **Thanh-Tho Le:** Resources. **Quoc-Hung Nguyen:** Resources. **Thuy-Chung Kieu-Le:** Methodology, Investigation, Resources. **Emilie Strady:** Conceptualization, Methodology, Validation, Writing – review & editing, Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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