Microplastics Occurrence in Surface Waters and Sediments in Five River Mouths of Manila Bay

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Microplastics have been increasingly documented globally in numerous environmental compartments. However, little information exists in the Philippines despite the fact that the country is considered to be one of the largest contributors of plastics in oceans. This study, considered as one of the pioneering microplastic research, evaluated the abundance, distribution, and composition of microplastic pollution in the mouths of five rivers, namely Cañas, Meycauayan, Parañaque, Pasig and Tullahan, draining to Manila Bay. Surface water and sediments samples were collected, then passed through a stack of sieves with sizes from 2.36 mm at the top to 0.075 mm at the bottom. These samples were digested to remove organic matter, and salt solutions were added to allow the microplastics to float. Extracted particles were examined under a stereo microscope, and quantified and categorized into shape, size, color, and type. Results show that microplastics were present ubiquitously at all river mouths but with concentrations varying from 1,580 to 57,665 particles/m^3 (surface water) and 386 to 1,357 particles/kg (dry sediment). Fragment was the most abundant shape, while white, blue, and transparent were the most prevalent colors. Fourier Transform Infrared Spectroscopy (FTIR) analysis revealed that polypropylene (PP), high and low-density polyethylene (high-density polyethylene and low-density polyethylene) and polystyrene were the main types of microplastics present in the river mouths.

Keywords: Manila bay, microplastics, plastic waste, sediment, surface water

INTRODUCTION

Microplastics are recently considered as an emerging pollutant, and these are divided into primary and secondary (Andrady, 2011; Wang et al., 2019). Primary microplastics are found in personal care products, cosmetics, medicines, and textiles as well as those by-products released from the use of plastic-related products such as tyre dust from running cars (Browne et al., 2011; Eriksen et al., 2013; Li et al., 2017; Wang et al., 2019; Suresh et al., 2020). Secondary microplastics are derived from the fragmentation of larger plastic and synthetic materials by processes such as UV degradation or machine washing (Andrady, 2011; GESAMP, 2015; Antunes et al., 2018). Microplastic contamination was first studied by Carpenter et al. (1972). Since then, many studies have documented the occurrence of microplastic contamination among the different bodies of water such as oceans (Anderson et al., 2016; Browne et al., 2011), estuaries (Browne et al., 2010; Lima et al., 2015; Zhao et al., 2014) freshwater bodies (Biginagwa et al. 2016; Free et al., 2014; Sanchez et al, 2014), and even in the remote arctic ice (Zarfle and Matthies, 2010; Hubard et al., 2014).

There is hardly any monitoring of the microplastic contamination in the Philippines despite a study by Jambeck et al. (2015) that in 2010, the country has been identified as the third largest contributor of plastic wastes in the ocean globally and that about 1.01 million tonnes of plastic waste is mismanaged by the country in 2016 (Law, et al., 2020). Within the last 5 years, studies on microplastics contamination in the Philippines have been published which reported the occurrence of microplastics in surface water (Argota et al., 2018; Espiritu et al., 2019; Lumongsod and Tanchuling, 2019), marine sediments (Bucol et al., 2019; Espiritu et al., 2019; Esquinas et al., 2020; Kalnasa et al., 2019), and marine biota such as fishes (Bucol et al., 2019; Espiritu et al., 2019; Paler et al., 2021), mussels (Argamino and Janairo, 2016), and oysters (Espiritu et al., 2019). However, Galarpe et al. (2021) mentioned that there is a need for more macroplastic and microplastic research in the country to develop and support appropriate plastic regulation policies.

This research presents an overview and serves as a benchmark for the initial report on the microplastic contamination in the surface waters and sediments of the Manila Bay particularly in the mouths of its tributary rivers namely Cañas River, Meycauayan River, Parañaque River, Pasig River, and Tullahan River. One of the country's widely used, semi-closed inlet for various economic, industrial, and domestic activities of more than 16 million Filipinos, Manila Bay, is surrounded by densely populated areas including Metro Manila (PEMSEA and MBEMP TWG RRA, 2004). Due to intensive anthropogenic activities and mismanaged plastic waste and dumping within the watershed of the bay, the surface waters and sediments of the bay may accumulate high concentration of microplastics.

It is important to gather baseline information to minimize health and environmental risks posed by these microplastics in the future, specifically to the water quality of marine and freshwater environment, marine biota and even to human consumption and health.

MATERIALS AND METHODS

Study Area

Manila Bay (about 220 km coastline and 17,000 km^2 drainage area) is a semi-enclosed marine inlet and its coastal water connects to the West Philippine Sea through a 16.7 km wide opening and surrounded by the National Capital Region and the municipalities of Cavite, Bulacan, Bataan, and Pampanga (Chang et al., 2009; Sia et al., 2009). It has 26 catchment areas including the river basins of Pasig and Pampanga, and a volume of approximately 31 km^3 with an estimated depth of 17 m on an average (PEMSEA and MBEMP TWG-RRA, 2004). The bay is primarily used for international and local port and harbor, fishing ground, aquaculture, and other maritime activities (Prudente et al., 1994).

At present, it has deteriorating water quality because of intensified disposal and mismanagement of solid and human wastes (Chang et al., 2009; Sia et al., 2009). Despite the fact that a large amount of solid wastes was drained into the bay from the highly populated and industrialized area of Metro Manila and its surrounding provinces, data is lacking on the microplastic pollution in the bay. These solid wastes, specifically the plastic wastes would degrade into smaller pieces due to biological, chemical, and mechanical action, thus contributing to the microplastics in Manila Bay.

The study mainly focuses on the quantification and characterization of microplastics extracted from the five major river mouths namely Cañas River, Meycauayan River, Parañaque River, Pasig River, and Tullahan River draining to Manila Bay (**Figure 1**). These sampling areas were selected as these are the major river systems traversing through the National Capital region and the adjacent cities, which drain to Manila Bay. The mouths of these rivers are also accessible. Location and description of the study areas are presented in **Table 1**.

Collection of Samples

All samples were collected from October 2018 to March 2019. Sampling was conducted once at each site with both surface water and sediments collected on the same day. Surface water samples were collected prior to sediments to avoid collecting suspended solids from the bottom of sampling sites. Water samples of 0.2 m^3 with about 5 m away from the riverbanks were collected using a bucket from 0 to 12 cm below the surface of different points in the sampling area. The use of bucket for surface water sampling is also used by Su et al. (2016), Argota et al. (2018), Lumongsod and Tanchuling (2019) and Suresh et al. (2020) as an alternative to the manta net. The use of manta net was not applicable in the study areas due to the presence of macroplastics and large debris along the rivers.

Water from the bucket was poured unto the stacked stainless sieves with mesh sizes of 2.36-mm (No. 8), 1-mm (No. 18), 500-μm (No. 35), 250-μm (No. 60), 125-μm (No. 120) and 75-μm (No. 200). Particles that are greater than 5 mm that were retained from the 2.36-mm sieve were discarded. The used sieves were thoroughly rinsed with distilled water and left to dry for an hour. The retained particles in each sieve were transferred to their respective glass containers using steel tweezers and a brush.

For sediment sampling at each of the five sites, three sediment samples, were gathered using an Ekman grab sampler (~0.1 m^2) and a scoop with minimal disturbance. Two of these were collected from bank deposits about 10 m away from each other while the remaining one sample was taken from the main channel deposits of the river, The topmost 5 cm of the sediments was collected, stored in separate glass containers, and transported to the laboratory for processing together with the surface water samples.

Overall, there were five surface water samples and 15 sediment samples collected from five river mouths, with six subsamples each representing the particles retained in mesh sieves giving a total of 120 subsamples to be processed for digestion, density separation and filtration.

Extraction of Microplastics

The extraction of microplastics was conducted in accordance with previous studies (Peng et al., 2017; Su et al., 2016; Zhang, 2017), with some minor modifications. All samples from the
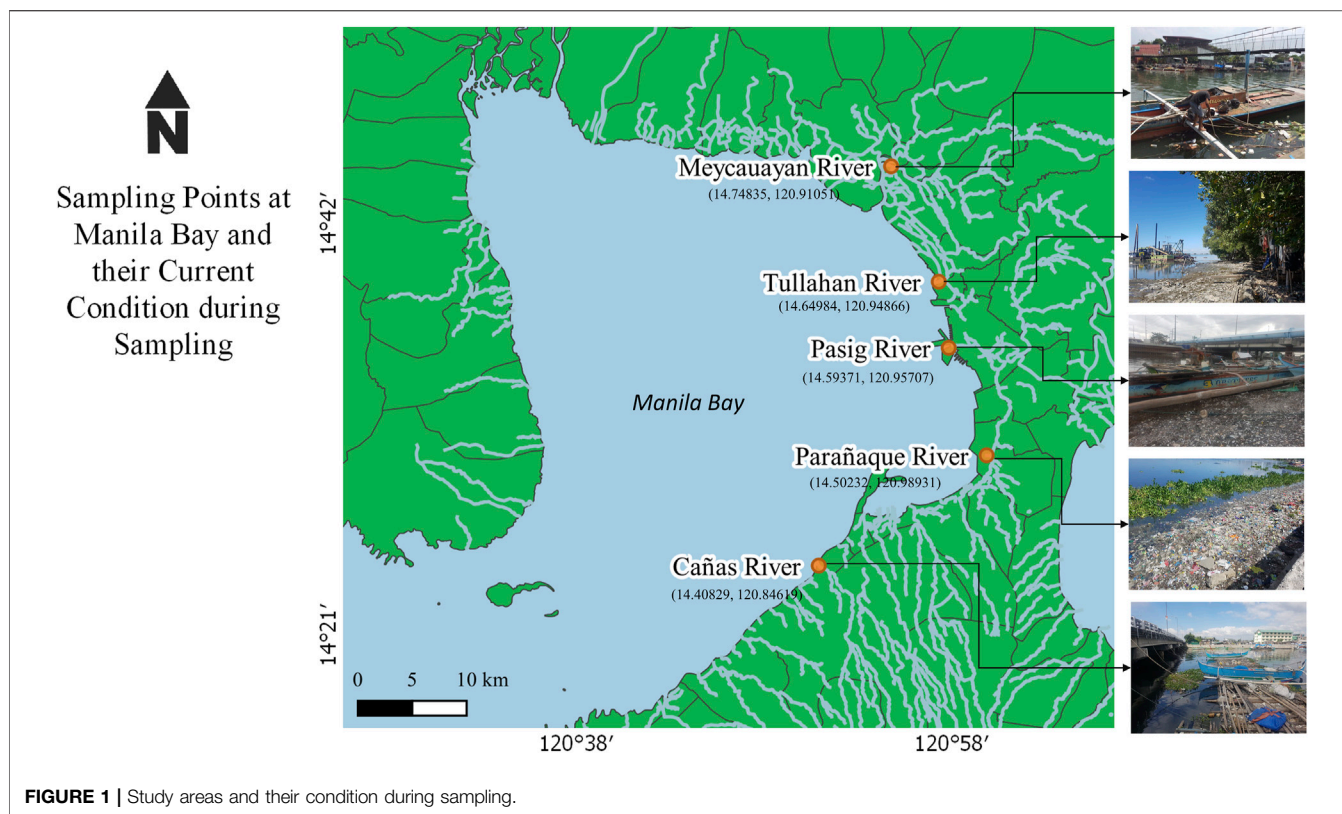
FIGURE 1 | Study areas and their condition during sampling.

TABLE 1 | Location and description of the study areas.

Area	Latitude	Longitude	Description of the study area
Cañas River	14°24′29.84″N	120°50′46.28″E	In Province of Cavite; Along the river are residential homes mostly informal settler families with a population of 50,717 (2017)
Meycauayan River	14°44′54.06″N	120°54′37.84″E	In Province of Bulacan; One of the worst polluted rivers; Presence of plastic manufacturing industries within the watershed of the river system
Parañaque River	14°30′8.35″N	120°59′21.52″E	In Metro Manila; Mouth of the river is beside the Las Piñas-Parañaque Critical Habitat and Ecotourism Area (LPPCHEA), the only natural wetland sanctuary for birds in Metro Manila, and a seafood market.
Pasig River	14°35′37.36″N	120°57′25.45″E	In Metro Manila; One of the worst polluted rivers; Mouth of the river is surrounded by the Manila port and Baseco Compound, a relocation site for informal settler families with a population of 59,847 (2015)
Tullahan River	14°38′59.42″N	120°56′55.18″E	In Metro Manila; Mouth of the river is surrounded by several facilities such as residential houses, elevated highways, bus terminals, pumping station and bancas and canoes; Presence of plastic manufacturing industries within the watershed

surface waters and sediments were oven-dried for 72 h with a temperature of 90°C.

After drying, an aliquot of 70 g in each sediment sample was taken and poured through a stacked arrangement of 2.36-mm (No. 8), 1-mm (No. 18), 500-μm (No. 35), 250-μm (No. 60), 125-μm (No. 120) and 75-μm (No. 200) stainless mesh sieves. Sediments that passed through the 75-μm sieve were discarded. The sediments in each sieve were transferred to their respective glass containers.

Wet peroxide oxidation (WPO) method was conducted to remove the organic matter present in all samples. At room temperature, 20 cm^3 of both aqueous 0.05 M Fe (II) solution and 30% H_2O_2 solution were added to the beaker containing collected solids in each size fraction (Yonkos et al., 2014; Masura et al., 2015). The samples were heated to 75°C and agitated at 200 rpm using a magnetic stirring hotplate for at least 45 min (Ling et al., 2017). In instances where natural organic materials were still visible, an additional 20 cm^3 of 30% H_2O_2 was continually poured to the mixture until no gas bubbles were observed.

After all the visible organic matter were removed, density separation method was applied which allowed the high-density polymers to float. High-density sodium chloride (NaCl; 1.202 g/cm^3) was added to the mixtures of the samples (Galgani et al., 2013). NaCl solution is an effective medium that allows plastics such as PE (0.91–0.97 g/cm^3), PP (0.94 g/cm^3), PS (1.05 g/cm^3),

TABLE 2 | Classification and categorization used for the collected microplastics.

Parameter	Categories
Shape	Fiber/line (elongated, fibrous, straight)
	Film (thin, soft, transparent)
	Fragment (hard, jagged, angular pieces)
	Foam (lightweight, porous, sponge-like)
	Pellet (hard, spherical, ovoid)
	Sheet (irregular flat, flexible)
Size	75–<125 μm
	125–250 μm
	250–<500 μm
	500–<1,000 μm
	1–<2.36 mm
	2.36–<5 mm
Color	White
	Transparent
	Red
	Orange
	Yellow
	Green
	Blue
	Black
	Other Color

and PVC (1.14 g/cm^3) to float (Van Cauwenberghe et al., 2015). However, for sediment samples of 75 microns in which the floating particles could no longer be seen by the naked eye, zinc chloride (ZnCl$_2$; 1.6 g/cm^3) was added to ensure the separation of all common polymers (Van Cauwenberghe et al., 2015). The samples were mixed on a hot plate with a magnetic stirrer at 300 rpm for about 15 min and were kept overnight covered with watch glass to observe clearance level wherein the microplastics floated and the other heavier particles settled down.

The supernatant water from the samples was transferred to another glass container, and then filtered using a Longer medium-high flow rate peristaltic pump with 47-mm Whatman® GF/C filters (pore size of 1 μm) to separate the microplastics.

Identification and Quantification of Microplastics

The extracted microplastics were examined and photographed using a 3.1-Megapixel CMOS camera connected to a ST-7045 Stereo microscope with magnification of 10x to 40x. The length and area of microplastics were analyzed using the ToupView software equipped with the digital microscope calibrated to a standard.

All the particles were sorted according to their color, size, and shape (depends on physical characteristics) using the classification from previous studies (Free et al., 2014; Li et al., 2016) as indicated in **Table 2**.

Polymer make-up was determined using FTIR spectroscopy (Nicolet 6700 FTIR ATR spectrometer with a diamond accessory). Extracted particles were grouped and representative microplastics in each group of at least 30 particles per site were chosen to test polymer types. This was done as a compromise between labor and cost which was also done by Peng et al., 2017.

Due to size constraints of ATR-FTIR Spectroscopy, it should be noted that only particles with sizes of 500 μm and above were analyzed. However, it is assumed that those particles with sizes below 500 μm which were subjected to wet peroxide oxidation and density separation are microplastics.

At a resolution of 4 cm^{-1}, thirty-two scans were collected in each sample, and each generated spectrum was documented in the 4,000–400 cm^{-1} spectral range. The attained spectra were analyzed using the BioRad KnowItAll® Informatics System (2018) software resourced with KnowItAll FTR spectral library that contained an intensive database of known compounds (Horton et al., 2017). For the reliability of data, only those spectra with matching score of more than 70% with the standard database were accepted (Zhao et al., 2018).

Quality Assurance and Quality Control

Usage of plastic containers for storage samples was avoided since these may cause additional microplastics in the samples, especially those that are not visible to the naked eye. The containers were properly labelled, stored in a box and transported to the laboratory within the same day for processing. Personal protective equipment like laboratory gowns, safety masks and nitrile gloves were always worn by the investigators during the experiment and analysis.

Collected solids in the glass containers were transferred to a clean beaker using minimal rinsing with squirt bottle containing distilled water, and the beaker was immediately covered with a watch glass to avoid spillage and contamination. Samples stored in glass Petri dishes were always covered to lessen the period of exposure.

The beaker and all the transfer apparatus were washed with distilled water multiple times to minimize any sample loss owing to adhesion of microplastics on the walls of the filter apparatus, and all washing solutions were filtered through the same glass-fiber filter. Filters were air dried and subsequently sealed individually in Petri dishes.

The crystal of the ATR-FTIR was cleansed using Kimtech Science Kimwipes and EtOH (96%) before and after analyzing each particle. Before the start of every FTIR analysis, a blank background scan was tested.

RESULTS AND DISCUSSION

Abundance of Microplastics

Microplastics were ubiquitously extracted in both surface water and sediment samples. The abundance of these particles, as illustrated in **Figure 2**, varied from 1,580–57,665 particles/m^3 in surface waters and 514–1, 357 particles/kg in dry sediments.

There is no uniform standard for recording the abundance of microplastics in surface water and sediment worldwide, hence not all the results from published studies can be used for comparison. In this case, only related studies that used similar quantification units (particles/m^3 for surface water; particles/kg of dry weight for sediment) for the abundance of microplastics were gathered as shown in **Table 3**. Even though these comparisons might be inaccurate because of the differences in the concerned particle

FIGURE 2 | Microplastic abundance in surface waters and sediments of five river mouths in Manila Bay.
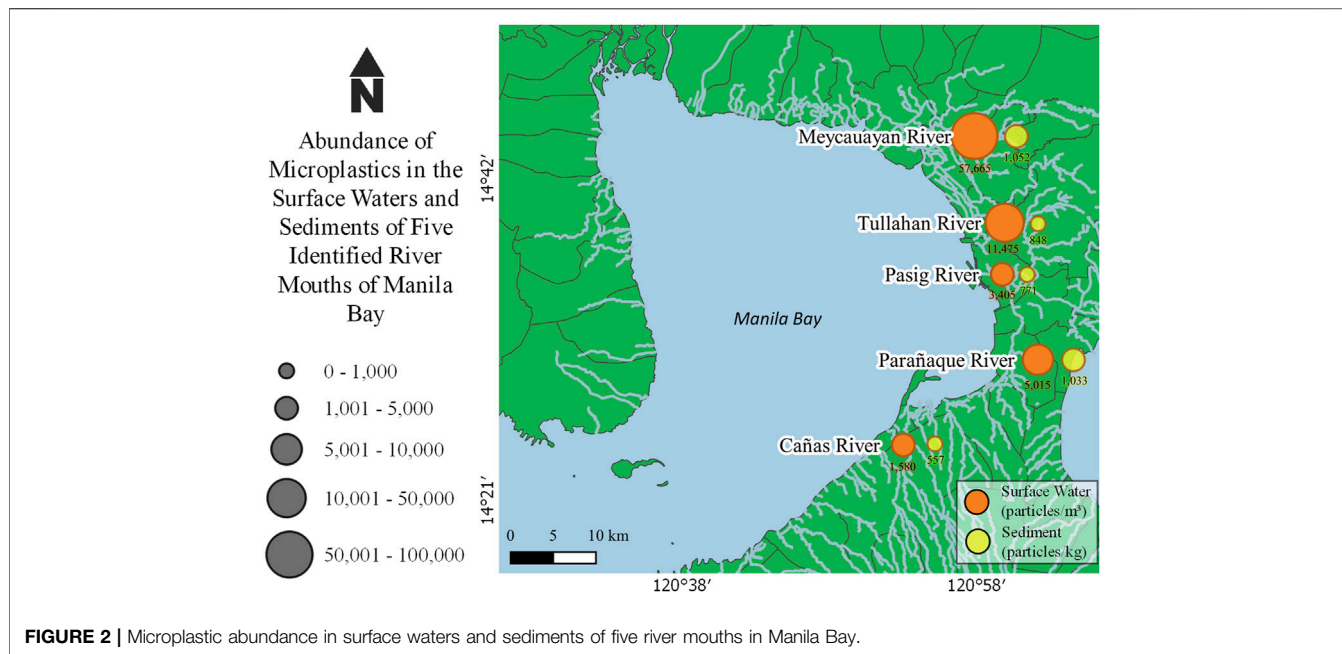
size being studied, sampling methodologies, and time and date of sampling, these studies still provide the general overview of the microplastic pollution worldwide (Tanchuling and Osorio, 2020).

Comparison of microplastic particle abundance in surface water across the globe with this study (**Table 3**) indicates a wide variation from as low as 0.73 particles/m³ in Lake Victoria of Uganda (Egessa et al., 2020) to as high as 30, 800 particles/m³ in the Winyah Bay of USA (Gray et al., 2018). Generally, the results in this study were relatively higher compared to other studies (Desforges et al., 2014; Zhao et al., 2014; Tsang et al., 2017; Simon-Sánchez et al., 2019; Egessa et al., 2020; Pan et al., 2020; Suresh et al., 2020). As for the Philippine setting, the abundance of microplastics in the surface waters of Meycauayan River and Tullahan River were at higher levels as compared to Tunasan River in Laguna, Lawaye River in Batangas, and creeks in Makati City (Argota et al., 2018; Espiritu et al., 2019; Lumongsod and Tanchuling, 2019).

With regards to the abundance of microplastics in the sediments, the sampling areas had higher levels than previous studies (Ng and Obbard, 2006; Claessens et al., 2011; Liebezeit and Dubaish, 2012; Van Cauwenberghe et al., 2013; Laglbauer et al., 2014; Nor and Obbard, 2014; Zhao et al., 2018) including those studies from the Philippines (Bucol et al., 2019; Espiritu et al., 2019) (**Table 3**).

Characteristics of Microplastics

Microplastics are existing in environmental compartments as particles having a variety of colors, shapes, sizes, polymer make-up and even densities (Zhou et al., 2020; Isaac and Kandasubramanian, 2021). These properties were found to be related with the environmental effects and fate of microplastics in the environment (Lambert et al., 2017) and even to the sinking of floating microplastics (Ryan, 2015; Kowalski et al., 2016).

Therefore, the extracted microplastics were classified and quantified according to these properties as shown in **Figure 3**.

Shape and Polymer Type

Microplastics were classified according to fragment, film, pellet/granule, line/fiber, sheet, and foam (**Figure 4**). These shapes could indicate the parent materials of the microplastics (Zhang et al., 2018).

Fragment as illustrated in **Figure 3** is the most dominant shape across all samples. Possible origins of these particles are from the degraded plastic products which include plastic containers, packaging materials and cleaning media (Derraik, 2002; Thompson et al., 2004; Zhang et al., 2015). Most of these fragments were identified as polypropylene (PP) which was the most abundant polymer type identified in most of the representative samples for both surface water and sediment (**Figure 3**). The abundance of these fragments maybe attributed to indiscriminate waste dumping and mismanaged plastic wastes as evidenced by the amount of macroplastics observed during sampling.

Films were also significantly abundant in all samples especially in the rivers of Cañas and Pasig as the mouth of these two rivers were surrounded by large residential settlements wherein direct littering and rampant garbage dumping were observed. Most films were detected as low-density polyethylene (LDPE) (**Figure 3**). These films could possibly originate from plastic wrapping, bags and packing materials (Zhang et al., 2015) including single-use plastics, wrappers and sachets observed in each river mouth.

Primary microplastics such as pellets that are mainly used as raw materials for manufactured plastic products (Hidalgo-Ruz et al., 2012; McDermid and McMullen, 2004), were extracted in most of the samples for both surface waters and sediments, indicating the contribution from the industry. These pellets

TABLE 3 | Mean abundance of microplastics reported in surface water and sediments.

Mean abundance		Study area and country	Concerned particle size	Reference
Surface Water (particle/m^3)	0.73	Lake Victoria (Uganda)	0.3–5 mm	Egessa et al. (2020)
	3.5	Ebro Estuary (Spain)	0.005–5 mm	Simon-Sánchez et al. (2019)
	11	Deep Bay (Hongkong)	<5 mm	Tsang et al. (2017)
	17	East China Sea (China)	0.5–5 mm	Zhao et al. (2014)
	55	Tsing Yi (Hongkong)	<5 mm	Tsang et al. (2017)
	253.1	Zhangjiang Estuary (China)	0.330–5 mm	Pan et al. (2020)
	279	Northeast Pacific Ocean (Canada)	<5 mm	Desforges et al. (2014)
	751.7	Cochin Estuary (India)	0.25–5 mm	Suresh et al. (2020)
	833	Makati Creeks (Philippines)	0.3–5 mm	Lumongsod and Tanchuling (2019)
	1,580	Cañas River (Philippines)	0.075–5 mm	This study
	3,333	Lawaye River (Philippines)	0.363–5 mm	Espiritu et al. (2019)
	3,405	Pasig River (Philippines)	0.075–5 mm	This study
	4,173	Yangtze Estuary (China)	0.5–5 mm	Zhao et al. (2014)
	5,015	Parañaque River (Philippines)	0.075–5 mm	This study
	7,547	Tunasan River (Philippines)	0.3–5 mm	Argota et al. (2018)
	7,630	Queen Charlotte Sound (Canada)	<5 mm	Desforges et al. (2014)
	8,902	Pearl Estuary (China)	0.050–5 mm	Yan et al. (2019)
	9,300	Haihe River (China)	0.333–5 mm	Liu et al. (2020)
	11,475	Tullahan River (Philippines)	0.075–5 mm	This study
	30,800	Winyah Bay (United States)	0.063–2 mm	Gray et al. (2018)
	57,665	Meycauayan River (Philippines)	0.075–5 mm	This study
Sediment (particle/kg)	1.8	Beach (Germany)	<1 mm	Liebezeit and Dubaish (2012)
	8	Beach (Singapore)	1.6 µm–5 mm	Ng and Obbard (2006)
	36.8	Mangrove (Singapore)	1.6 µm–5 mm	Nor and Obbard (2014)
	72	South Yellow Sea Offshore (China)	1 µm–5 mm	Zhao et al. (2018)
	75	Lawaye River (Philippines)	0.045–5 mm	Espiritu et al. (2019)
	82	North Bais Bay (Philippines)	<2 mm	Bucol et al. (2019)
	123.6	North Yellow Sea Offshore (China)	1 µm–5 mm	Zhao et al. (2018)
	144	Beach (Canada)	<5 mm	Ballent et al. (2016)
	166.7	Harbor (Belgium)	0.038–1 mm	Claessens et al. (2011)
	171.8	Bohai Sea Offshore (China)	1 µm–5 mm	Zhao et al. (2018)
	177.8	Beach (Slovenia)	0.25–5 mm	Laglbauer et al. (2014)
	557	Cañas River (Philippines)	0.075–5 mm	This study
	760	Tributary (Canada)	<5 mm	Ballent et al. (2016)
	771	Pasig River (Philippines)	0.075–5 mm	This study
	848	Tullahan River (Philippines)	0.075–5 mm	This study
	980	Nearshore (Canada)	<5 mm	Ballent et al. (2016)
	1,033	Parañaque River (Philippines)	0.075–5 mm	This study
	1,052	Meycauayan River (Philippines)	0.075–5 mm	This study
	1,340	Cochin Estuary (India)	0.25–5 mm	Suresh et al. (2020)
	1,423.5	Subtidal (Italy)	0.7 µm–1 mm	Vianello et al. (2013)
	31,050	Tidal Flat (Germany)	<1 mm	Liebezeit and Dubaish (2012)

were identified as high-density polyethylene (HDPE) and could originate from the spillage and personal care products (Fendall and Sewell, 2009; Eerkes-Medrano et al., 2015) (**Figure 3**). Interestingly, higher number of pellets were extracted from surface waters of Meycauayan River (particle count of 204) compared to the other rivers being studied (particle count of less than 35). Possible origin would be leakage from the plastic manufacturing industries within the watershed of Navotas-Malabon-Tenejeros-Tullahan and Marilao-Meycauayan-Obando River System.

Lines and fibers were also identified in the samples for all sites. These were identified as PP, PE and PET. PET was only identified in the surface water sample of Cañas River while PP and PE were present in all sampling sites (**Figure 3**). These were probably fragment of plastics originating from fishing nets, ropes, plastic straws and even textiles (Cole et al., 2011; Wang et al., 2017). Aside from garbage dumping and degradation of larger plastics,

washing is also considered as an important pathway that releases these lines, fibers, and other airborne particles into the environment (Dris et al., 2016; Salvador Cesa et al., 2017). Numerous fishing activities, aquaculture, and harbors for canoes and bancas present in the sampling areas could be considered as main contributors to the presence of lines and fibers in the samples. The most abundant type in the sediment samples of Parañaque River was line. It is likely due to the existence of large amount of plastic straws, ropes, and fishing nets from the canoes and bancas docked beside the seafood market along the mouth of the river.

Foams mostly polystyrene (PS) were sufficient in amount in the surface waters especially in the rivers of Meycauayan and Tullahan, wherein thousands of foams were extracted (**Figure 3**). These particles were also identified in the sediments of all study sites in low quantities. It is observed that concentration of PS in surface waters were more abundant than those in the sediments.
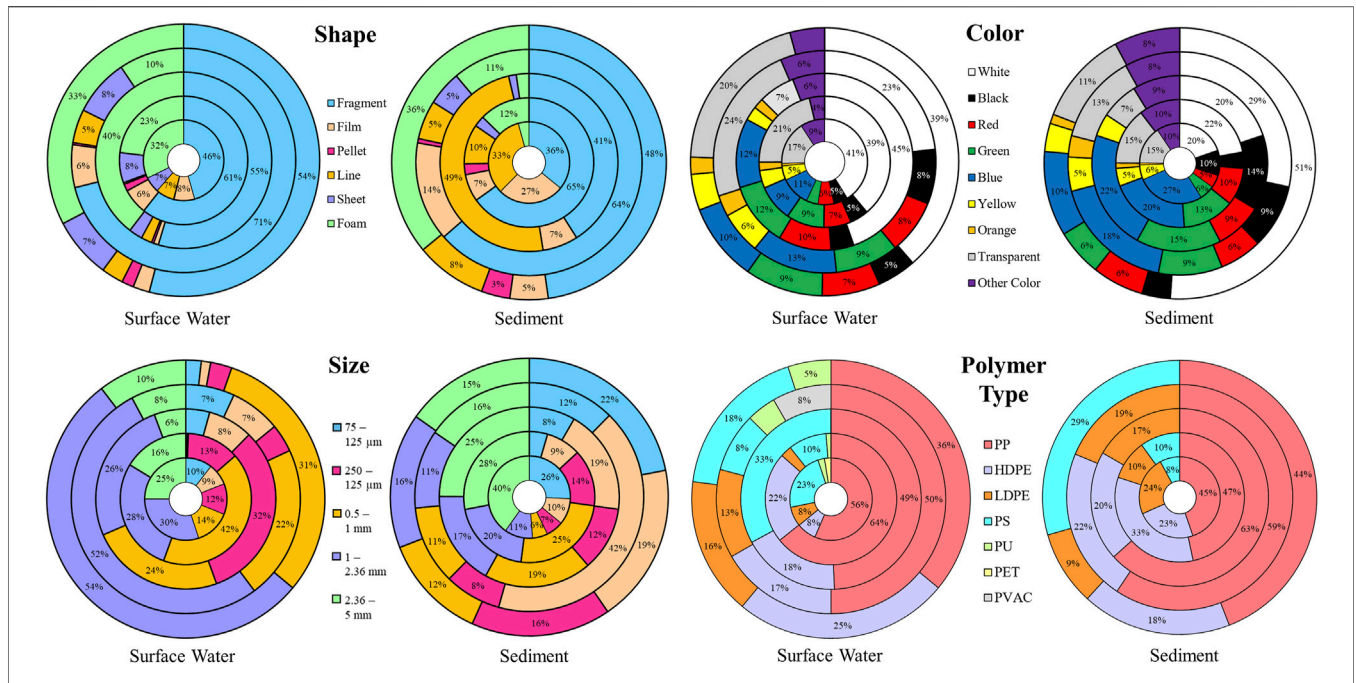
FIGURE 3 | Distribution of characteristics of microplastics in surface waters and sediments of five river mouths in Manila Bay (from innermost to outermost circle) – Cañas, Meycauayan, Parañaque, Pasig, and Tullahan.
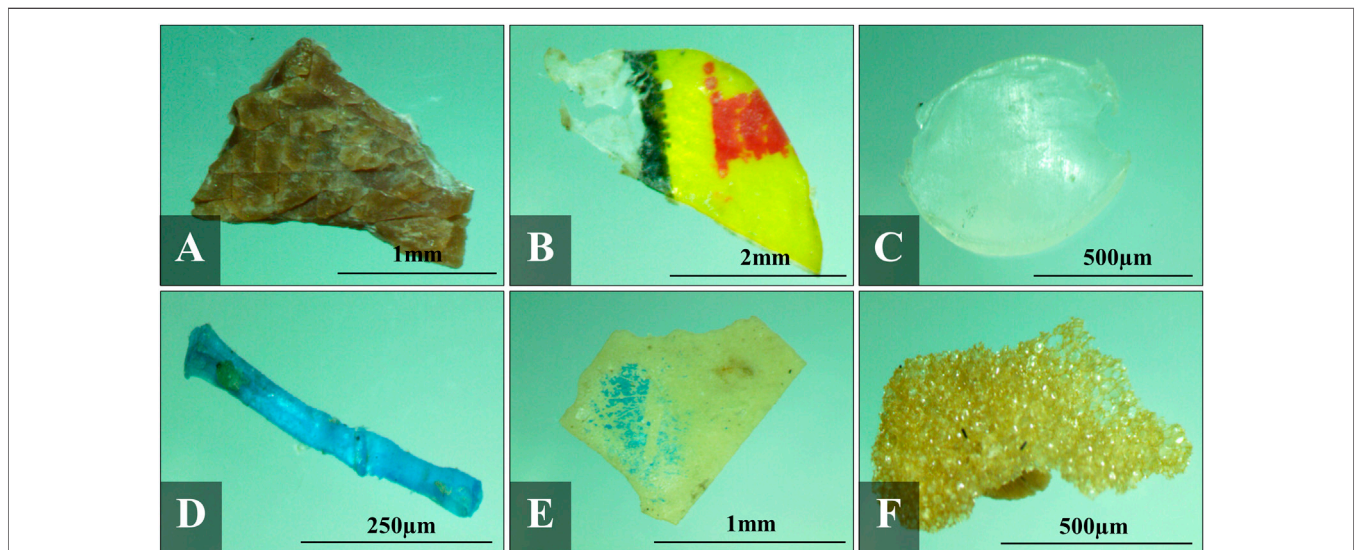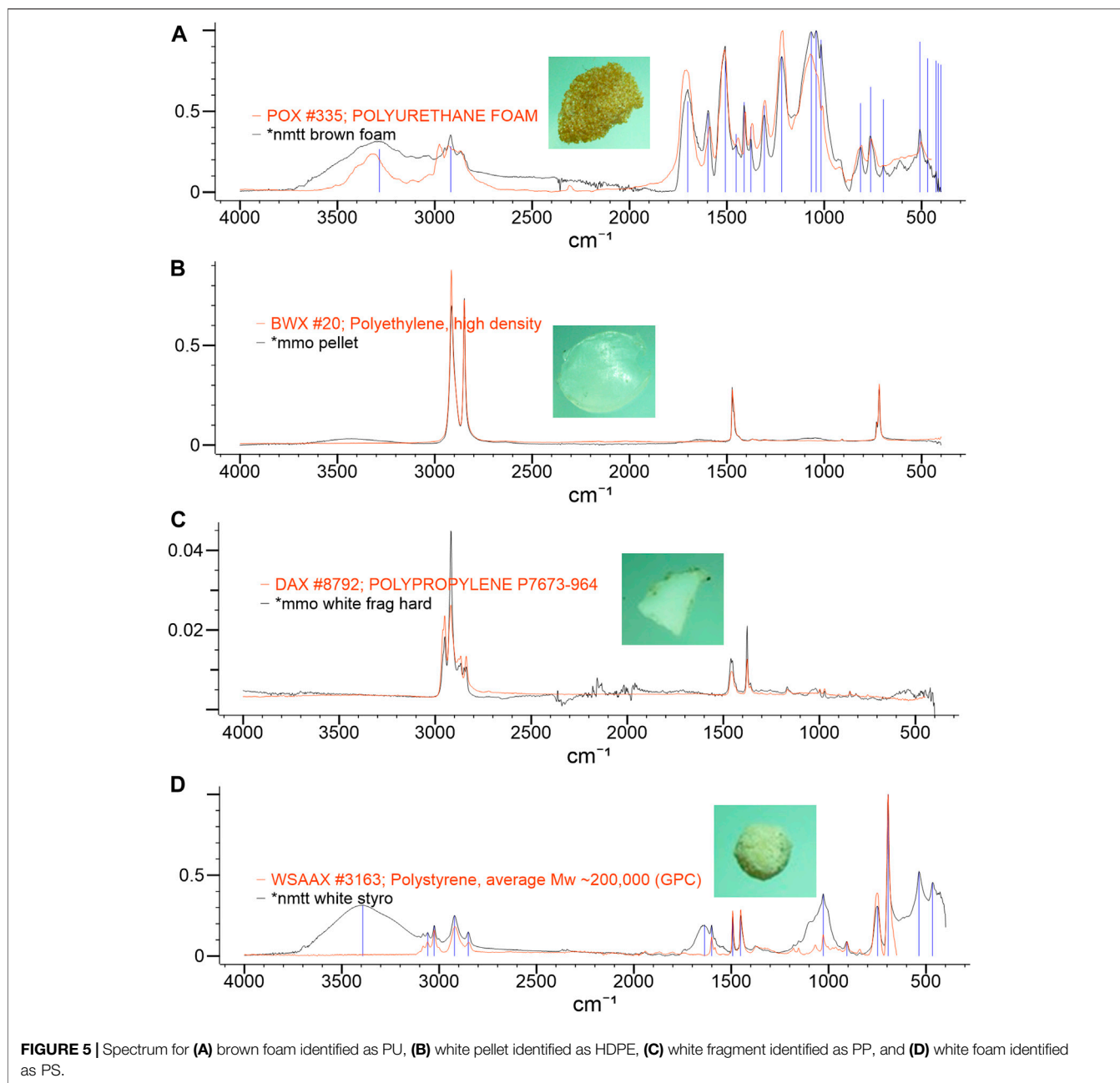

FIGURE 4 | Photographs of different shapes of microplastics collected in all samples: **(A)** fragment, **(B)** film, **(C)** pellet, **(D)** line, **(E)** sheet, and **(F)** foam.

This is likely due to the air injected into the foam, specifically Styrofoam, making it less dense and allowing it to float. Aside from PS, polyurethane (PU) was identified in some foams in the surface waters of Cañas, Meycauayan, Pasig and Tullahan River, but in low quantities only. Site visit and ocular inspection on the sampling areas determined that the possible origin of the foams present in these sites were from large quantities of disposable plastic containers produced such as food and product containers

utilized for food delivery services and takeout food, and even from the foam fishing floats and rafts used by fishermen (Di and Wang, 2018).

Significant number of sheets was detected, and these particles were also associated to the degradation of larger plastic products especially for those macroplastics existing in the sampling areas (Cole et al., 2011). Analyzed sheet particles were mostly PP and HDPE. One of the sheets in the surface water sample of Pasig

FIGURE 5 | Spectrum for **(A)** brown foam identified as PU, **(B)** white pellet identified as HDPE, **(C)** white fragment identified as PP, and **(D)** white foam identified as PS.

River was identified as polyvinyl acetate (PVAC) which is primarily used as adhesives for porous materials such as white glue. It is possible that the adhesive material stuck on the particle was identified by ATR-FTIR instead of the polymer make-up of the sheet itself.

Low-density polymers including PP, PE, PS and PU particles are expected to float in surface water because their densities are lower as compared to freshwater. However, the polymer types of PP and PE were the most identified types of plastic among the analyzed samples in the sediments, implying that density is not the only factor affecting the microplastic distribution in the surface waters and sediments. These low-density particles could also be conveyed through current and deposited on the

sediments through the following mechanisms: 1) increase in net density of microplastics through biofouling (Andrady, 2011), 2) natural substances adsorption to the surface (Frias et al., 2010), 3) inclusion of inorganic fillers during manufacturing (Corcoran et al., 2015), and 4) fecal express (Cole et al., 2015).

Interestingly, PET whose density is higher than the freshwater was detected in the surface waters of Cañas River even though it is expected to settle down, although in relatively smaller amounts. This might be due to environmental factors such as temperature, wind driven turbulence, tides and waves, these denser plastic particles could have been resuspended from the deeper waters to the surface (Zhao et al., 2014). PET bottles were commonly
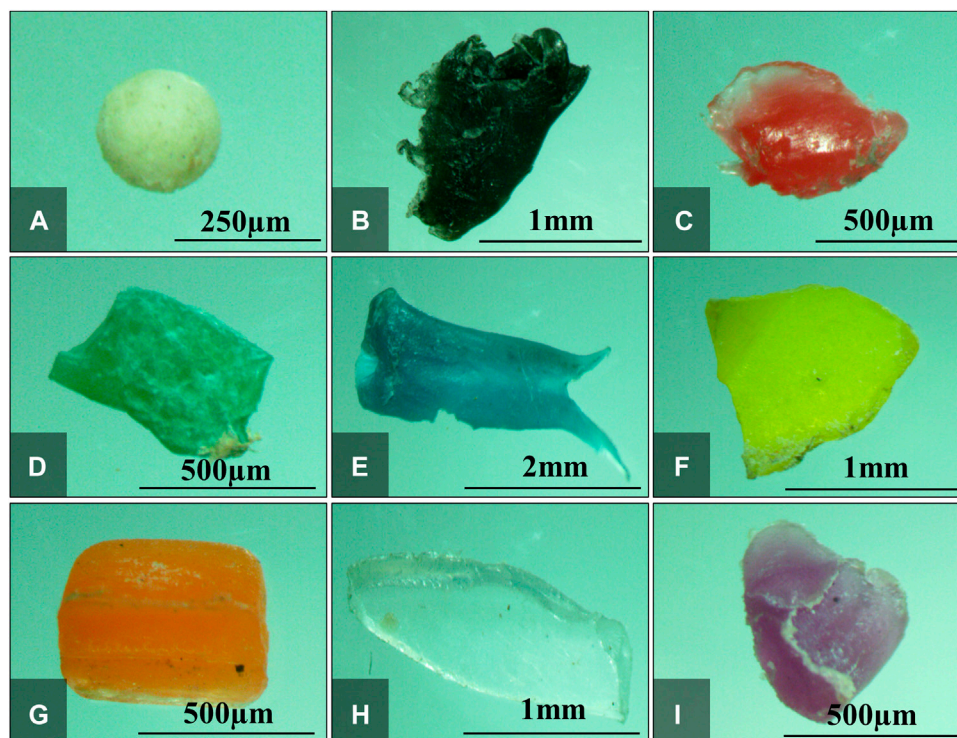
FIGURE 6 | Photographs of different colors of microplastics collected in all samples – **(A)** white, **(B)** black, **(C)** red, **(D)** green, **(E)** blue, **(F)** yellow, **(G)** orange, **(H)** transparent, and **(I)** violet.

collected and sold to junkshops; hence this might be one of the reasons why PET particles were barely identified in the samples.

Spectra were generated for all the analyzed particles. Some of these are shown in **Figure 5**, and the additional spectral band observed may be associated to the plastic additives or the adsorption of persistent organic pollutants or organic materials (Cole et al., 2011). Further investigation is recommended to identify these specific substances adsorbed to the microplastic samples.

The abundance of the fragments, films, lines and fibers, foams and sheets suggested that majority of the collected microplastics in the mouths of the rivers being studied were locally derived secondary microplastics (Wessel et al., 2016; Yonkos, et al., 2014).

The observed morphological patterns such as scratches, creases, micro-pores, and cracks, among others in the extracted microplastic particles indicate that their source from the degradation of products such as plastic containers, packaging materials, and ropes (Zhang et al., 2016). Moreover, the shape of microplastics were modified by different forces such as the actions from tides and waves (Karthik et al., 2018). Several factors including origins, debris quality, breakdown of plastics, and plastic sinking rate could control the plastic distribution, hence diverse shape distributions in the sampling areas were identified (Critchell and Lambrechts, 2016).

Color

The microplastic particles were also classified into colors such as red, orange, yellow, green, blue, black, white, transparent, and other colors including violet, brown, and multi-colored particles.

Photographs of typical microplastic samples categorized into different colors of microplastics were presented in **Figure 6**.

White colored particles may represent the color of plastics such as Styrofoam™, cap bottles, disposable spoon and fork, and ice cream cups while transparent colored microplastics may come from the virgin pellets, cellophanes, single-use plastics, plastic packing bags and boxes used as food containers. Other identified color particles such as red, green, blue, yellow, orange, violet, and brown were possibly fragmented from commonly used products such as packaging, toys, household products, sachets, plastic straw ropes, and food wrappers. Colored microplastics detected in the environment could be related to the high consumption of the colored plastic products in everyday living (Zhang et al., 2015; Wang et al., 2017).

Color could also be one of the factors for the ingestion of microplastics by marine biota due to resemblance of their prey (Wright et al., 2013). Based on previous studies, it was found out that fish may mistakenly feed on microplastics with colors of white, brown, and yellow that most closely resemble their zooplankton prey (Shaw and Day, 1994; Boerger et al., 2010). Moreover, it was discovered that translucent and light-colored plastics were commonly ingested by sea turtles (Bugoni et al., 2001) and some shapes and colors of plastics were accidentally ingested by some seabirds (Barboza and Gimenez, 2015).

As illustrated in **Figure 3**, white was the most prevalent color among all surface and sediments samples. This color was also the most observed in Guanabara Bay (Castro et al., 2016) and Northeast Atlantic Ocean (Lusher et al., 2013). These white particles primarily consisted of foams, pellets, and fragments.
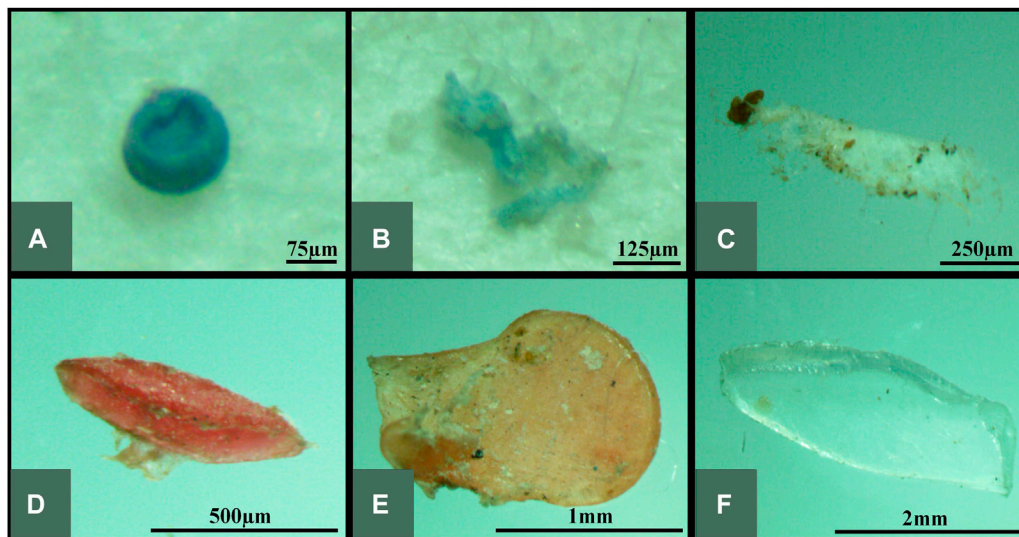
FIGURE 7 | Photographs of different sizes of microplastics collected from the surface water and sediment samples – **(A)** 75 – <125 µm, **(B)** 125 – <250 µm, **(C)** 250 – <500 µm, **(D)** 500 – <1000 µm, **(E)** 1 – <2.36 mm, and **(F)** 2.36 – <5 mm.

Most of the plastic pellets extracted from the surface waters and sediments were white and transparent, like previous studies conducted (Turner and Holmes, 2011; Corcoran et al., 2015). This finding is expected since white and transparent pellets are the most common color manufactured (Redford et al., 1997). However, there were very few amounts of black, blue, and orange pellets extracted in some samples, which was also observed by Young and Elliott, 2016.

Transparent and blue were also identified abundantly in all samples. These transparent particles were recognized to be dominant in surface water samples and possibly originated from the short-lifetime and disposable packaging plastic products such as containers, bottles, cups and bags. Furthermore, blue microplastics were mainly comprised of fragments and lines (most probably fishing lines and plastic straws). High prevalence of blue-colored microplastics in the aquatic environment could increase the likelihood of ingestion since some marine fishes reportedly ingested blue microplastics as food (Güven et al., 2017; Ory et al., 2017). Moreover, blue microplastics were also frequently found in clams (Shi, 2018).

Other colored microplastics such as red, green, and yellow were also found in significant amount in the samples as these could be derived from different plastic packaging materials (Zhang et al., 2018). High abundance of colored plastics, especially colored fragments, attributed to the broad usage of coloration in the manufacturing, since coloration is used to attract consumers of plastic products (Thetford et al., 2003).

Size
It has been reported that microplastic ingestion is primarily determined by size and abundance as compared to color and it

occurs by chance (Rodríguez-Sejio and Pereira, 2017). The microplastics could pose risks to the marine biota as demonstrated in fish larvae wherein organisms likely to consume their prey with the smallest dimension (Hunter, 1981). These particles are now considered available for ingestion to aquatic organisms since the sizes of these approximate the size of some planktons (Cole et al., 2011; Wright et al., 2013; Cole et al., 2015). However, due to the differences in the results from published research, further studies are needed to verify which among the attributes of microplastics aid the ingestion.

The extracted particles in this study were classified into six size categories as shown in **Figure 7**. The lower size limit of microplastics in this study is 75 µm. Among these different size categories of microplastic particles, the class 1–2.36 mm, contributed the highest quantity in the surface water samples in most of the study sites, except in the rivers of Meycauayan and Parañaque wherein the most abundant size category was 500–1,000 µm and 250–500 µm, respectively (**Figure 3**).

In general, about 80% of total microplastics recovered in the samples from surface water were within the size class of 250 µm–5 mm. The size range of these microplastics were still considered visible to the naked eye.

In terms of the microplastic size extracted from the sediment samples, it is observed in **Figure 3** that the sizes were relatively evenly distributed in each study site as compared to the distribution of sizes of microplastics collected in the surface water samples. Microplastic particles with size class of 2.36–5 mm, contributed the highest quantity in the sediment samples in most of the study sites, except in the rivers of Pasig and Tullahan wherein the most abundant size category was 125–250 µm and 75–125 µm, respectively. Despite the differences in the percentage distribution of sizes, a significant amount of microplastics was extracted from each size class in each study site.

Source, Fate and Transport of Microplastics

The highest concentration of microplastics for both surface waters and sediments were found to be in Meycauayan River while the lowest was detected in Cañas River (**Figure 2**). All sampling was done during ebb tide wherein axial convergence occurs near the bottom of the river mouths and axial divergence prevails at the surface making floating matters such as microplastics are pushed towards the banks (Wolanski and Elliot, 2015). This could explain the occurrence of microplastic particles collected in the surface water and sediment along the banks in the mouths of the river sampling.

It could be observed that from south (Cañas River) to north (Meycauayan River), the concentration of microplastics in surface waters tend to increase (**Figure 2**). The abundance of microplastics in the rivers of Pasig, Tullahan and Meycauayan could be further correlated with the calculated plastic emissions of these rivers from the study of Meijer et al. (2021). This could also be associated with the presence of several plastic manufacturing industries within the watershed of Marilao-Meycauayan-Obando and Navotas-Malabon-Tenejeros-Tullahan River Systems. This is supported by the presence of pellets which are considered as primary microplastics.

With regards to the wind transport in the bay, individual average wind blow at specific period of the year-controlled bay's circulated gyres differently as simulated by De Las Alas and Sodusta (1985). There are Northeasterly winds, with speeds averaging about 5 m/s from October to March and Southwesterly winds, with speeds of 5–7 m/s from April to September. Since all sampling were conducted during Northeast monsoon, the effect of winds could possibly contribute to the high abundance of microplastics collected in the rivers of Meycauayan and Tullahan as the direction of the flow of the winds is in counterclockwise direction. While the direction of winds at the southeastern part of the bay where the rivers of Parañaque and Cañas are located is in clockwise direction, the microplastics at their river mouths might be flushed out to the bay.

However, it is recommended to conduct more detailed studies on the combined influence of river runoff, tidal input, coastal currents, and wind transport to the abundance of microplastics in Manila Bay.

In terms of the concentration of microplastics in the sediments, the microplastic pollution levels in sediments from the bay were more closely related to the distance from the contamination source (Su et al., 2016) and these microplastic most observed in Guanabara B particles could only be highly disturbed, collected and transported during dredging activities. For example, the amount of microplastics extracted in the sediments in Parañaque River could have possibly originated from the seafood market beside its river mouth.

Based on the study of Siringan and Ringor (1998), the transport direction of surface sediment is southward off Pampanga and northeastward off Cavite. The bottom sediments are always transported from the bay mouth into the bay, and the sediments do not flow from Manila Bay. This would imply that the sediments were more stable as compared to surface water, and the suspended microplastics in the sediments will be transported slower than those floating in surface water (Su et al., 2016).

Manila Bay has a narrow mouth and a relatively weak tidal current, hence the residence time of water inside the bay is relatively longer (Pokavanich and Nadaoka, 2006). It is possible that the microplastics floating in the surface water in each river mouth will be drained off into the bay and will be retained in the bay for a long period of time. These floating microplastics could retain and undergo degradation and be biofouled, making them dense enough to sink (Andrady, 2011; Van Cauwenberghe et al., 2013). This scenario would likely increase not just the concentration of microplastics in the surface waters, but also in the bottom sediments of Manila Bay.

CONCLUSION

The widespread occurrence in different environmental compartments of microplastics has been recorded worldwide, however, few studies are available on the microplastics present in the aquatic systems in the Philippines. This is the first extensive microplastic study in Manila Bay. This bay serves as an important area for fishing ground and aquaculture activities, hence the occurrence of microplastics in this bay will likely impact on marine organisms and the entire food web through bioaccumulation.

Surface water and sediment samples were collected in the river mouths of Cañas, Meycauayan, Parañaque, Pasig and Tullahan which are draining to Manila Bay. The abundance of microplastics ranges from 580 to 57,665 particles/m^3 in surface water and 386 to 1,357 particles/kg in dry sediment. The levels of microplastic pollution found in these rivers are currently at higher levels, compared to the other studies. Secondary microplastics were found to be the most abundant as fragments indicating that these originate from plastic wastes which were improperly disposed. Therefore, prevention of leakage of both macroplastics and microplastics in the tributary rivers and the bay itself is very important to preserve the marine environment in Manila Bay.

More extensive studies in different waterbodies, and other environmental compartments with sampling in both wet and dry periods to account the effect of seasonal aberration in the abundance and distribution of microplastics are recommended to provide a clear overview on the extent of microplastic pollution in the country. The rehabilitated areas and other tributaries draining to Manila Bay are suggested to be used as sampling areas for further studies to provide more assessment regarding the microplastic contamination in the bay.

DATA AVAILABILITY STATEMENT

The raw data supporting the conclusions of this article will be made available by the authors, without undue reservation.

AUTHOR CONTRIBUTIONS

EO, MT and MD conceived of the idea, developed the methods, and edited the manuscript. EO conducted the experiment and ran analyses. All authors approved the submitted version of the manuscript and agreed to be listed.

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