

The abundance, characteristics and diversity of microplastics in the South China Sea: Observation around three remote islands

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HIGHLIGHTS

- A high abundance of floating MPs was found in the southern South China Sea.
- Transparent film and fiber were predominant in water and organisms, respectively.
- 84.7% of floating MPs and 54.5% of MPs in vivo belonged to PP and PE.
- Characteristics of MP in organisms were different from those of inshore ones.

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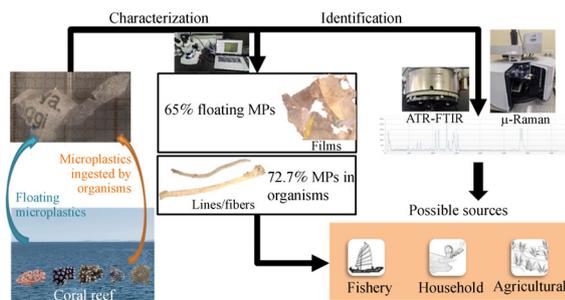
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ABSTRACT

Surrounded by emerging markets with considerable plastic consumption, the South China Sea has been a focus area of microplastic research. A survey on the floating microplastics (> 0.3 mm) and microplastics ingested by fish and mollusks was conducted around three remote islands here. Compared with the results from several previous studies, a high abundance of floating microplastics (with a median of 1.9×10^3 items/km² or 0.7 items/m³) was observed, revealing another “hot spot” for microplastics. Polyolefin, especially polypropylene, was the main component. The diversity index and evenness index were calculated and evaluated based on the composition of microplastics. The characteristic peaks of Raman spectra concerning pigmented microplastics were provided. Transparent sheets/films were predominant in the water sample, which was quite different from a similar study in this sea area (8.9% for film), and only 16.4% of floating microplastics (> 0.3 mm) were fibers/lines, implying that the main sources of floating microplastics (> 0.3 mm) might be household/agricultural consumption activities. The transparent fiber/line was also dominant in organisms. It is suggested that the main sources of microplastics ingested by organisms might be both fabric fibers and fishing/aquaculture.

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1 Introduction

Microplastics (MPs), an omnipresent pollutant in the marine environment, have been found in the remotest (Suaria et al., 2020) and deepest parts of oceans (Peng

et al., 2018). Based on their sources, MPs are classified into primary MPs (i.e., those intentionally produced or appear in the natural environment in small sizes; Enfrin et al., 2019; Hidayaturrahman and Lee, 2019) and secondary MPs (i.e., those degraded from larger plastics). Based on an estimate (Eriksen et al., 2014), there were 4.85×10^7 items or 35,540 tons of MPs floating globally in the oceans. Considering that this prediction also noted that 233,400 tons of plastics > 5 mm floated on the sea surface, the abundance of MPs may increase rapidly.

Several main economies encompassing the South China Sea (SCS), including China, Indonesia, the Philippines,

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Vietnam, Thailand and Malaysia, are among the estimated top 10 economies of plastic waste inputs from land to sea (Jambeck et al., 2015). Based on the Morgan Stanley Capital International Emerging Market Index, having been or being about to become (Vietnam) emerging markets, these economies are facing rapid consumption growth and subsequent per capita waste production growth. In addition, Malaysia, Indonesia were still top importers of plastic waste after the former leading economy (China) dramatically reduced its imports (WTO, 2020). As estimated by Borrelle et al. (2020), once the improvement of their waste management system lags behind this growth trend, the amount of waste entering aquatic ecosystems, particularly plastic waste, is likely to increase greatly (Lestari and Trihadiningrum, 2019). In fact, some previous studies have reported high mean levels of floating MPs in coastal areas around the SCS, such as 2.4 items/m³ (> 333 μ m) in the Pearl River Estuary (Harris et al., 2021), 421.8 items/m³ (> 20 μ m) in the Terengganu Estuary (Taha et al., 2021), and 4.5 items/L (> 20 μ m) in the Maowei Sea (Zhu et al., 2019b).

Coral reefs are an important ecosystem in the SCS. High biodiversity is observed there, especially around remote islands such as the Nansha Islands. Two hundred species of scleractinian (reef-building) corals (approximately two-thirds of the world species) have been found in the Nansha Islands, with 6500 marine species recorded around the atolls (Liu, 2013a). There has been evidence suggesting that both scleractinian corals (de Carvalho-Souza et al., 2018) and coral reef species (such as sea anemones and coral reef fish; Jensen et al., 2019; de Orte et al., 2019) could ingest plastics. Although the negative effects of MPs on the health of coral species are still controversial (Berry et al., 2019; Reichert et al., 2019), exposure to MPs has been proven to increase the stress response of scleractinian corals (Reichert et al., 2018; Tang et al., 2018b; Lanctôt et al., 2020). Several types of plastic additives detected in reef-building corals were also supposed to be derived from MPs (Saliu et al., 2019; Montano et al., 2020). Given that MP exposure might also pose a potential threat to planktivorous reef fish (Critchell and Hoogenboom, 2018), it is of great significance to investigate the MPs suspended around coral reefs. To date, MPs in seawater have been investigated around several islands of the Xisha Islands (Ding et al., 2019) and Nansha Islands (Huang et al., 2019; Nie et al., 2019; Wang et al., 2019; Tan et al., 2020) and in the open sea of the SCS (Cai et al., 2018). For a variety of considerations, the MPs in most of these studies were mainly collected with pumps, water samplers or bongo nets, which could not provide information about the surface MPs (GESAMP, 2019). Considering that in general, the highest concentration of suspended MPs occurs on the surface (Reisser et al., 2015), a survey with a Neuston net trawl can supplement the relevant information. Surface-trawl data sets have been applied to build a global floating plastic model (van Sebille et al., 2015). In

addition, since MPs collected in surface trawl samples could be much more abundant than those collected from pumped samples per site (Tamminga et al., 2019), they are suitable for MP diversity evaluation. Based on the characterization of MPs, new insights into the source apportionment of MPs might be provided by MP diversity evaluation (Wang et al., 2019).

MPs ingested by field collected organisms living around coral reefs in the SCS have also been investigated (Ding et al., 2019; Nie et al., 2019; Tang et al., 2021). MPs could be selectively enriched in corals and other organisms (Tang et al., 2021). The impact of MPs on marine organisms is supposed to be affected by the bioavailability and physiochemical properties of MPs (e.g., size and density; Wu et al., 2019). The size of MPs ingested is affected by the main food of the related organism (Moore and Phillip, 2011), and their intake might depend on the living habit of the organism (Wu et al., 2019). For example, pristine low-density plastics might be ingested by organisms living in the upper water column, whereas benthos are more likely to encounter dense plastics. Therefore, the types and sizes of MPs ingested by organisms are often different from those in surface water. The monitoring of MPs in biological samples could reveal the characteristics of MPs exposed to local communities.

Considering that a supposed high abundance of MPs in surface seawater and various possible sources of MPs (and different MP diversity from other sea areas) in this sea area, the aims of this study were to: 1) investigate the abundance and characteristics of MPs in surface water- and field-collected organisms in warm-water coral reef ecosystems in the shallow seas of the SCS; 2) assess the MP diversity in the study area; and 3) compare the composition of MPs between this sea area and other sea areas.

2 Materials and methods

2.1 Study area

Meiji Reef (Mischief Reef; 115°32'E, 9°55'N), Chigua Reef (Johnson South Reef; 114°17'E, 9°42'N) and Huayang Reef (Cuarteron Reef; 112°50'E, 8°51'N) are the parts of Nansha Islands and are located in the southern SCS (Table S1). The areas of these three atolls are estimated to be 46 km², 7 km², and 8 km² by satellite images, respectively. No industrial or agricultural activities on or around the three reefs have been approved or planned except for Meiji Reef (according to media reports, there is small-scale aquaculture in the south-eastern part of the lagoon in Meiji Reef).

2.2 Sample collection

Ten surface water samples were collected from May 22 to June 3, 2019 (six samples around Meiji Reef, two samples

around Chigua Reef, and two samples around Huayang Reef), following a similar Neuston net trawl method reported previously (Chen et al., 2019; Pan et al., 2019). Specifically, samples were collected with a Neuston net (0.6 m × 0.4 m rectangle mouth; 0.333 mm mesh; ~4 m long) at an average trawling speed of approximately 2 km/h for a period of 15 min. The net was set on the side of the vessel during trawling, keeping the angle of ~20° between shipping routes. The initial net immersion depth was ~0.2 m in still water. The trawling distance is estimated by a flow meter (Chen et al., 2019). The sampling area is estimated as the product of trawling distance and net width (Table 1). A stainless steel collector is attached to the bottom of the net. At the end of each trawling, the net was lifted at a speed of approximately 0.5 m/s and rinsed with pumped surface seawater onsite from the outside of the net to drive all the solids inside the net down to the collector. And then the sample in the collector was transferred to a 1-L glass vial. The water depths at all of the sampling stations are > 3 m.

All of the biological samples (Table S2) were collected in the waters around Meiji Reef. Due to the prohibition of commercial fishing activities in coral reef areas, organism samples were obtained by divers. After collection, the organisms were wrapped with aluminum foil and stored at -20°C before further treatment.

2.3 Sample treatment

All Neuston samples were treated in a modified manner recommended by Masura et al. (2015), which has been applied previously (Pan et al., 2019). Organism samples were treated in the same manner described previously (Fang et al., 2019).

Briefly, Neuston samples in the sampling vial were poured through stacked stainless steel mesh sieves with mesh sizes of 5.0-mm and 0.3-mm, respectively. The residues on 5.0-mm sieves were rinsed repeatedly with Milli-Q water to avoid adhesion or entanglement of smaller solids (including MPs). After rinse with Milli-Q water to remove salt and solid < 0.3 mm, all the material on the 0.3-mm sieves were then transferred into glass beakers (with scale and glass covers), and dried in the oven at 75°C. 20 mL hydrogen peroxide (H₂O₂, 30%) and 20 mL of 0.05 mol/L Fe(II) aqueous solution for each beaker was first added. The suspension was heated at 75°C until bubbles appeared, and then the beaker was removed from the hotplate. After bubbles disappeared, the suspension was kept at 75°C and stirred for approximately 0.5 h. If the solution in the beaker was still not clear, another H₂O₂ or Fe (II) solution would be added as appropriate, and repeated the above process to make sure that the natural organic matters were fully oxidized. The suspension was filtered through a glass fiber filter (Whatman GF/A, 1.6 µm pore size) using a vacuum system. Milli-Q water was used

in the rinse of filter at last, to remove the salt on the filter. Finally, all solids on the filter were collectively placed in a clean petri dish covered with aluminum foil and air-dried overnight.

The total length and weight of each individual of organism samples was recorded (Table S2). Immediately after that, the individuals were rinsed with filtered Milli-Q water, and dissected. The whole gastrointestinal tracts (GIT) of fish and the entire soft tissues of mollusks were sampled and weighed. Subsequently, 5 mL of 10% KOH solution per gram (wet weight) sample was added to digest the samples for 6 h at 60°C and 300 rpm in an oscillation incubator. Afterwards, saturated NaCl solution was added in the digestion suspension with 1:1 by volume, setting overnight in density separators. The collection and preservation of solids in overlying suspension was similar that described in Neuston sample treatment. During filtration, the filtered Milli-Q water was heated to dissolve lipids.

2.4 Characterization

Solid substances collected were observed, classified, photographed and recorded according to their apparent characteristics, such as size (expressed as maximum Feret's diameter < 0.3 mm, 0.3–0.5 mm, 0.5–1 mm, 1–2.5 mm and 2.5–5 mm; Michida et al., 2019), shape (fiber/line, fragment, sheet/film and foam) and color (transparent, white, yellow, red, blue, green, black and others). The suspected MPs were identified and characterized by a Senterra II micro-Raman spectrometer (µ-Raman; Bruker Optics Inc., Billerica, MA). An optical microscope with 20 × and 50 × objectives (Infinity, USA) was equipped. All the MP samples were excited with 785 nm diode lasers of 1–100 mW. Raman spectra were recorded as random measurements (3–5 points) on various parts of the focused items to avoid interference from impurities. Suspected MPs that could not be identified by µ-Raman were analyzed with attenuated total reflection Fourier transformed infrared spectroscopy (ATR-FTIR; Bruker Hyperion 3000) with a 20 × ATR objective. This process is based on the fact that in general, for MPs within the range of 10–500 µm, more items could be identified by µ-Raman than by µ-FTIR or ATR-FTIR (Cabernard et al., 2018), especially for PE. All Raman (FTIR) spectra with a frequency resolution of 4 cm⁻¹ (8 cm⁻¹) and range of 50–3650 cm⁻¹ (600–3800 cm⁻¹) were analyzed using OPUS 7.8 software (Opus software Inc., San Rafael, CA). Both commercial libraries and self-created library references of typical polymers (Table S3 and Fig. S1) were adopted. For Neuston samples, the composition of 703 suspected items (including 650 items with a size of 0.3–5 mm) was identified by µ-Raman or ATR-FTIR, and 645 items were plastics (including 594 MPs); for organism samples, 33 MP items were identified.

2.5 Quality assurance (QA) and quality control (QC)

The QA and QC processes were employed similar to those used previously (Fang et al., 2018; Pan et al., 2019). Briefly, to minimize potential contamination, all reagents were filtered with Whatman GF/A glass fiber membrane filters, and all of the containers, separating funnels, glass slides, tweezers and needles were pre rinsed repeatedly with Milli-Q water. The labware with/without samples kept covered with glass covers or aluminum foils except for the operation. Only cotton laboratory coats were worn during the processing, detection and identification of samples. During the sampling of floating MPs, surface water onsite was used to wash away the impurities outside the net. Before the first sampling and after the last sampling each day, the seawater used for the rinse of the net was pumped, went through the outside of the net, collected, and transferred to glass vials as blank samples (Michida et al., 2019; ten blank samples totally). Two positive controls (polyethylene with diameter between 300 μm and 355 μm , and polystyrene with mean diameter of 0.99 mm) containing three replicates were set up by spiking the known amounts (approximately 100 items) particles into Milli-Q water. For MPs in the gastrointestinal tract of organisms, each ~ 20 g of fish flesh was taken as a procedural blank (five samples totally), and processed synchronously during the whole course to monitor the background contamination. Four positive controls (polystyrene and polypropylene particles with diameters of 50 μm and 100 μm) were set up by spiking the known amounts particles into the fish flesh samples. The blank samples and positive controls were treated with the same procedure as samples.

Only some unknown impurities (suspected to be residues of biomass treated by hydrogen peroxide) were detected in the blank samples for Neuston net, and none were identified as MPs. The procedural blanks for organism samples contained 1.20 ± 0.84 items/filters suspected MPs, none of which were identified as MPs at last (Fig. S2). The recoveries in treated positive controls of water samples were $98.1\% \pm 1.0\%$ for polyethylene (PE) and $96.5\% \pm 0.8\%$ for polystyrene (PS); those in biological samples were $85.1\% \pm 4.0\%$ for 50 μm PS, $92.6\% \pm 7.2\%$ for 100 μm PS, $88.3\% \pm 2.7\%$ for 50 μm polypropylene (PP) and $96.1\% \pm 3.1\%$ for 100 μm PP.

2.6 Surface MP abundance correction

The surface MP concentrations were corrected to avoid the variations induced by wind mixing. Wind speeds were measured with an anemometer, and then the Beaufort numbers were deduced from average wind speeds during trawling. The modified Kukulka model described by Reisser et al. (2015) was used for this correction. The details are provided in the Supporting Information.

2.7 Stimulated surface current

Surface current velocities and directions were obtained from the CORA v1.0 regional ocean reanalysis system for the North-west Pacific (cora.nmdis.org.cn; spatial resolution: $0.5^\circ \times 0.5^\circ$, 35 layers), developed by the National Marine Data and Information Service of China (NMDIS).

A detailed introduction is provided in the Supporting Information.

2.8 Data analysis

The Simpson diversity index was used by Wang et al. (2019) to calculate MP diversity. Shannon–Wiener's diversity index (H') and Pielou's evenness index (J') were used to calculate the heterogeneity of beach litter (Munari et al., 2016). In this study, based on the composition of MPs, H' and J' were applied to compare the diversity and evenness of floating MPs. They could quantitatively evaluate how the compositions were different at each site and how evenly the relative abundances of MPs were distributed among those compositions. The components with fewer than three occurrences were counted as "others" and were not included in the MP diversity assessment.

One-way analysis of variance (ANOVA) was applied to determine the significance of the differences in MP diversity among this study and other open sea areas investigated in a similar manner (Pan et al., 2019; Wang et al., 2020). The variances of the data (H' and J') across the groups were homogeneous (Levene's test), and the data were close to normality (Shapiro–Wilk test). When differences were detected (at $p = 0.05$), a post hoc LSD test and a post hoc Bonferroni test were carried out to perform all pairwise comparisons between group means. When grouped by different reefs, the floating MP abundance data in this study were not normal and the sample amount in each group was small; thus, a Kruskal–Wallis H test was applied instead to test the differences among the different reefs. ANOVA, Kruskal–Wallis H test and relevant tests were performed by SPSS 16.0.

3 Results and discussion

3.1 The abundance of MPs

Before correction, the abundance of floating MPs ranged from 2.5×10^4 items/ km^2 to 1.4×10^6 items/ km^2 , with an average of $(3.0 \pm 4.1) \times 10^5$ items/ km^2 and a median of 1.8×10^5 items/ km^2 ; the corrected abundance of MPs ranged from 2.7×10^4 items/ km^2 to 2.4×10^6 items/ km^2 , with a mean value of $(4.6 \pm 7.0) \times 10^5$ items/ km^2 and a median of 1.9×10^5 items/ km^2 (Table 1). The mean value was an order of magnitude higher than the mean value estimated

by Eriksen et al. (2014) for the world's ocean, or our survey results for the North-west Pacific, the mid-west Pacific and the harbor (Xiamen Bay) in recent years (Table S4; Chen et al., 2019; Pan et al., 2019; Wang et al., 2020). One possible reason is the shorter trawling time compared with those in our previous survey, but it is more likely that there is another “hot spot” of MP pollution in the southern SCS, since 594 MPs were identified from the Neuston samples in this study. The mean and median abundance could also be estimated as 1.1 ± 1.5 items/m³ and 0.7 items/m³ (the estimation method was provided in the supporting information), respectively, equivalent to several tens of times those investigated around the Nansha Islands earlier (with a mean value of 0.0556 ± 0.0355 items/m³; Tan et al., 2020). The difference in specific locations in the SCS and the variance in surface currents between May and June (Fig. S3) could be possible reasons; different features (especially shape) of MPs found are another explanation (Section 3.3). As Goldstein et al. (2013) suggested that the MP abundance varied greatly at the submesoscale (tens of km) and between different surveys (with an order of magnitude higher in summer than that in fall), this

potential “hot spot” should be confirmed by more surveys.

The abundance of MPs ingested by organisms is shown in Table 2. The mean detection frequency of MPs in the GIT of fish samples was 56.3%, with an abundance of 1.00 ± 1.21 items/individual (0.26 ± 0.41 items/g-wet weight). The abundance of MPs in soft tissues of mollusks was 5.66 ± 5.51 items/individual (0.05 ± 0.05 items/g-wet weight), with a detection frequency of 66.7%. The MP abundances in the GIT of fish were much greater than those provided in previous studies around Nanxun Reef of the SCS (Nie et al., 2019) but close to the data collected in the open sea of the SCS (Chen et al., 2021; Table S5). Considering that this sea area is geographically close to the sea area around China, the MP ingestions by organisms obtained from the two sea areas were compared (Table S5). Generally, the MP ingestions by fish in this study were less than those detected in samples obtained from China's market (Zhu et al., 2019b), whether measured by weight or by individual. Similarly, the MP ingestion of *Trochus niloticus* (by weight) was less than that found in China's shellfish samples in previous studies (Table S5), although this result might also be due to the heavier soft tissue of

Table 1 Observed quantity and abundance of floating MPs (0.3–5 mm), and correlating coefficients at each station

Site	Number (items)	Area ^a (m ²)	Average wind speed (m/s)	Beaufort number	H_s^b (m)	N_{tow}^b		N^b	
						(items/km ²)	(items/m ³)	(items/km ²)	(items/m ³)
Meiji Reef									
1	66	397	2.1	2	0.2	166,429	0.6	167,908	0.6
2	17	140	2.1	2	0.2	121,847	0.5	122,930	0.5
3	124	395	4.8	3	0.6	314,090	1.2	630,100	2.9
4	17	70	3.5	3	0.6	241,600	0.9	394,851	1.8
5	180	124	3.5	3	0.6	1,448,503	5.4	2,367,317	10.6
6	35	118	3.5	3	0.6	295,535	1.1	482,998	2.2
Chigua Reef									
7	43	435	3.0	2	0.2	98,938	0.4	102,678	0.4
8	6	235	3.0	2	0.2	25,545	0.1	26,511	0.1
Huayang Reef									
9	25	329	1.5	1	0.1	75,964	0.3	75,964	0.3
10	81	400	1.5	1	0.1	202,362	0.7	202,362	0.7

a) The relevant formulas were reported previously (Chen et al., 2019); b) Described in Supporting Information.

Table 2 The detection ratio and abundance of MPs in soft tissues or gastrointestinal tracts (GIT) of different species

Species	Parts	Individuals	Detection ratio	Abundance	
				(items/g-wet weight)	(items/individual)
<i>Trochus niloticus</i>	Soft tissues	3	66.7%	0.05 ± 0.05	5.67 ± 5.51
<i>Cephalopholis argus</i>	GIT	6	50.0%	0.16 ± 0.18	0.50 ± 0.55
<i>Myripristis murdjan</i>	GIT	7	42.9%	0.30 ± 0.45	0.71 ± 0.95
<i>Priacanthus macracanthus</i>	GIT	1	–	0.27	1
<i>Sargocentron caudimaculatum</i>	GIT	1	–	1.57	4
<i>Scarus festivus</i>	GIT	1	–	0.18	3

Trochus niloticus. It means that inshore aquaculture activities and land-based pollution can improve the probability of the MP ingestions by fishes and shellfishes.

3.2 The distribution of floating MPs and the possible input via surface current

The MP abundance around Meiji Reef, Chigua Reef, and Huayang Reef was $6.9 \pm 8.4 \times 10^5$ items/km² ($1.2 \times 10^5 - 2.4 \times 10^6$ items/km²), $6.5 \pm 5.4 \times 10^4$ items/km² ($2.8 \times 10^4 - 1.0 \times 10^5$ items/km²), and $1.4 \pm 0.9 \times 10^5$ items/km² ($7.6 \times 10^4 - 2.0 \times 10^5$ items/km²), respectively. Although the average abundance around Meiji Reef was much higher than that of the other two reefs, the differences between groups were not significant ($p > 0.05$). This result implies that the differences between the mean abundance in the sea area around Meiji Reef and those around others are mainly from some extremely high values.

Densely populated coastal areas and runoff through these areas are regarded as the most important sources of plastics in the ocean (Jambeck et al., 2015; Lebreton et al., 2017). However, in remote sea areas far away from land-based sources, the transport and accumulation of floating plastics were also affected by hydrodynamic features. Floating plastics models based on surface current have been developed (Lebreton and Borrero, 2013; van Sebille et al., 2015). Large floating plastics that sit high above sea level are considered more likely to be affected by the additional wind stress, which could explain the deviation between the predicted accumulation zone and that observed (Lebreton and Borrero, 2013); for MPs, the effect of additional horizontal wind stress may not be important. Therefore, it could be assumed that the distribution of land-based MPs is mainly affected by surface currents. The study area was in the southern part of the SCS (8–10°N). As shown in the stimulated current (Fig. S3), during November and May, the sea area around Meiji Reef is mainly affected by currents from the eastern SCS, i.e., coastal currents from Luzon Island and Palawan Island and currents from the Sulu Sea; between June and September, currents from the Indochina Peninsula and Kalimantan Island are dominant. The waters surrounding Chigua Reef are controlled by the currents that flow by Kalimantan Island year round, except that they are also affected by currents of the eastern SCS from March to May. The sea area around Huayang Reef was affected by a mesoscale cyclonic eddy south of the reef in summer or a basin-scale circulation south of the SCS (Nansha circulation; Liu, 2013b) in winter, both of which flow by Kalimantan Island. In general, this sea area could be affected by the land-based sources around the southern SCS, which could be evidenced by some product labels (Fig. S4). Tan et al. (2020) also found product labels in various languages on macroplastics around the Nansha Islands, indicating potential sources including China, Vietnam, and the Philippines. However, it is difficult to

further determine the main sources of MPs during the investigation. First, secondary MPs were considered to be dominant in the size categories of $> 250 \mu\text{m}$, even downstream of wastewater treatment plants (Estahbanati and Fahrenfeld, 2016), which were viewed as one main source of primary MPs. In marine environments, it takes a relatively long time (Min et al., 2020) to form a certain amount of MPs from macroplastics. Second, even for Meiji Reef, which is closest to the potential land-based sources (~ 250 km from Palawan Island), nearly a week is needed to transport land-based MPs with the fastest current speed obtained by simulation. During this period, the distribution of MPs in the sea area might be governed by many complex processes (van Sebille et al., 2020); the constantly changing currents also make any precise traceability impossible. In addition, due to the influence of the monsoon, the west side of the sea area will be affected by large-scale current input from the north of the SCS (winter) and Java Sea (summer). To assess the impact of input from these areas, it is necessary to carry out similar investigations on these sea areas. Finally, sea-based sources (such as fisheries) could not be ignored (Tan et al., 2020).

3.3 Characteristics of MPs

Of all the collected floating MPs, the ratio of MPs in the size range of 1–2.5 mm was the highest (Fig. 1), which was similar to that in the mid-west Pacific Ocean (Wang et al., 2020). Regardless of how MPs were discharged into the marine environment, they were degraded at a slow speed and gradually became smaller. Given this, the fact that in this study, the higher percentages of MPs in larger size groups (1–2.5 mm and 2.5–5 mm) than those reported previously (Pan et al., 2019; Wang et al., 2020) implies that the mean retention time of MPs in this area may be shorter than those in the center of the Pacific Ocean.

Although it is suggested that fibers were only poorly retained in coarse nets (e.g., $\sim 300 \mu\text{m}$ in this study; Dris et al., 2018; Taminga et al., 2019), the proportion of fibers/lines in suspended MPs in the mid-west Pacific Ocean was up to 57.4% (Wang et al., 2020). In contrast, in this study, only 16.4% of MPs were fiber/line; rather, sheet/film was dominant (Figs. 2 and S4). This result indicates that there were different sources of MPs between these two areas. Another investigation around the Nansha Islands in a similar manner reported 8.9% film (Tan et al., 2020). Considering that only 82 suspected MPs were identified by μ -FTIR during that investigation (Tan et al., 2020), the difference between their reported abundance and the data provided by this study might also be attributed to the underestimated items of film-shaped MPs.

Of all the observed MPs ingested by organisms, the median size was smaller than that of MPs found in seawater (Fig. 1; not considering those smaller than

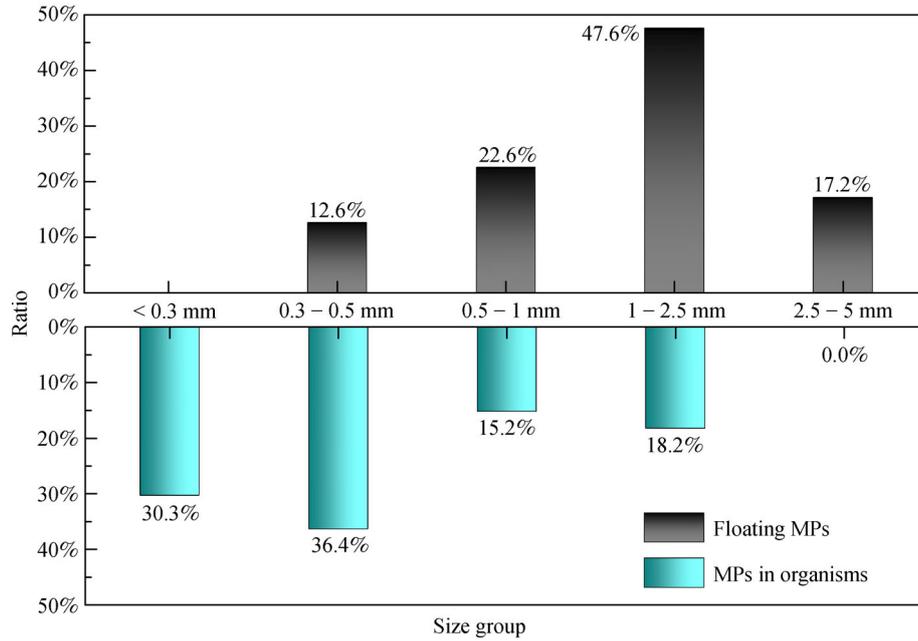


Fig. 1 The percentages of MPs in different size groups. None of the MPs ingested by organisms were found in the size group of 2.5–5 mm.

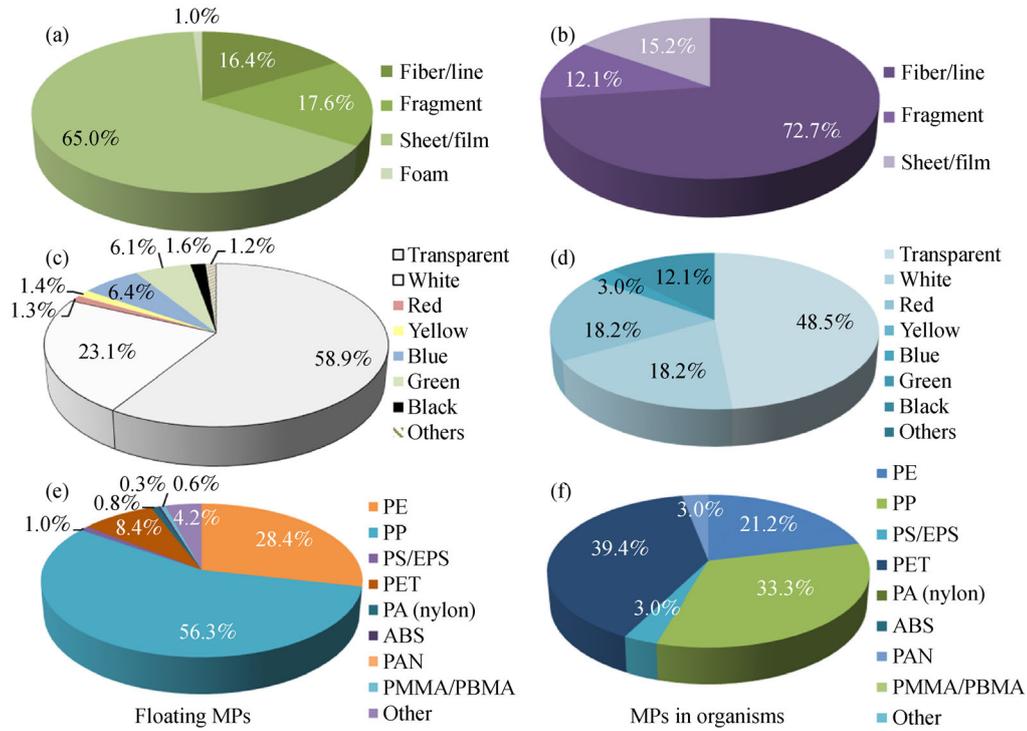


Fig. 2 The percentages of MPs with different shapes (a, b); different colors (c, d); different compositions (e, f). Pies (a), (c) and (e) represent the data of floating MPs; (b), (d) and (f) represent the data of MPs in organisms.

0.3 mm). Taking into account the size of their daily food ingested, it is not surprising. Fiber/line accounts for 72.7% (78.3% for MPs > 0.3 mm), higher than that in water (Fig. 2), but lower than that found in organisms of China’s coastal areas (Fang et al., 2019). Jensen et al. (2019) and

Ding et al. (2019) also suggested that coral reef fish did not randomly intake microdebris, the chemical composition, shape and color of which differ significantly from those detected in surface waters.

In both surface water and organisms, most MPs were

transparent, followed by white. The ratios of green, blue and red MPs were also not negligible, especially in those ingested by organisms (Fig. 2). The colors of plastics may be related to their sources (Shaw and Turner, 2019), but what makes more sense is that the colors might be important characteristics for assessing the potential ecological risk of MPs. Zhu et al. (2019a) proposed that seabirds on Yongxing Island of the SCS might mistake plastics as food items. In addition to the dimethyl sulfide (or other odors) signal from biofouling developing on plastic surfaces (Savoca et al., 2016; Dell'Aricecia et al., 2017), colors might be one of the suspected factors that lead to this misunderstanding for birds (Codina-García et al., 2013; Zhu et al., 2019a) or fishes (Sparks and Immelman, 2020). Considering that there are approximately 100,000 marine bird individuals and at least 3048 species of fish living in the SCS (Ma et al., 2008; Zhu et al., 2019a), providing information about the colors of MPs might help in forming an answer to the question of whether this color preference exists.

Some colored MPs could be confirmed as pigmented plastic particles with Raman spectra, providing information about their sources. Shim et al. (2017) suggested that Raman spectroscopy might suffer from interference from pigments; other studies pointed out the possible types of dyes or pigments with plastics or suspected plastics (Collard et al., 2015; Imhof et al., 2016; Karami et al., 2017; Zhao et al., 2017; Dowarah and Devipriya, 2019), such as copper phthalocyanine (CuPc). In this study, MPs (especially PP and PE) with various colors (blue, green, red, and black) were identified by Raman spectroscopy. Pigments with different colors show different characteristic peaks (Fig. 3). For MPs, the fluorescence background tends to be stronger at lower Raman shifts. Therefore, the Raman shift caused by inorganic pigments (e.g., titanium dioxide and chrome yellow) or metal ions of metal-complex dyes/pigments (e.g., Cu in CuPc) in MPs

might be covered by fluorescence, resulting in a weak signal (Fig. 3). However, some organic pigments show obvious peaks within the range of 1520–1610 cm^{-1} (1529–1533 cm^{-1} for blue pigments/dyes; 1594–1600 cm^{-1} for red pigments/dyes; 1534–1540 cm^{-1} for green pigments/dyes), which were not covered by the original peaks of MPs. These peaks of blue MPs might be assigned to C-N stretches arising from the CNC group (pyrrole) of H₂Pc (Marshall, 2010). The presence of H₂Pc could also be supported by other small Raman peaks (e.g., 683 cm^{-1} assigned to macrocycle breathing and 483 cm^{-1} assigned to isoindole ring deformation; Marshall, 2010). Metallization (such as CuPc or Pigment Blue 15; Denekamp et al., 2019) or the change in the groups connected to the CNC group (e.g., chlorinated CuPc or Pigment Green 7; Burgio and Clark, 2001) could slightly change the Raman shift of the C-N stretching. Some organic red pigments/dyes (e.g., naphthol red or Pigment Red 170; Caggiani et al., 2016) have also been found to generate strong signals in this range (1520–1610 cm^{-1}). However, not all characteristic peaks in this study were obvious. For example, PE with black color shows only a broad, low-intensity absorption peak at approximately 603–1596 cm^{-1} (corresponding to carbon black or Pigment Black 7; Caggiani et al., 2016); Cabernard et al. (2018) also found that black color MPs were harder to be identify than blue MPs with μ -Raman.

3.4 The main composition of MPs

A total of 84.7% of floating MPs were polyolefins; other compositions included PET (polyethylene terephthalate). The ratio of polyolefins was larger than that reported previously in adjacent sea areas (58.5%; Tan et al., 2020). Polyolefin (PE and PP) sheets/films are typical secondary MPs from grocery bags and food wrappers (Chen et al., 2019; Wang et al., 2019); polyolefin films are also believed

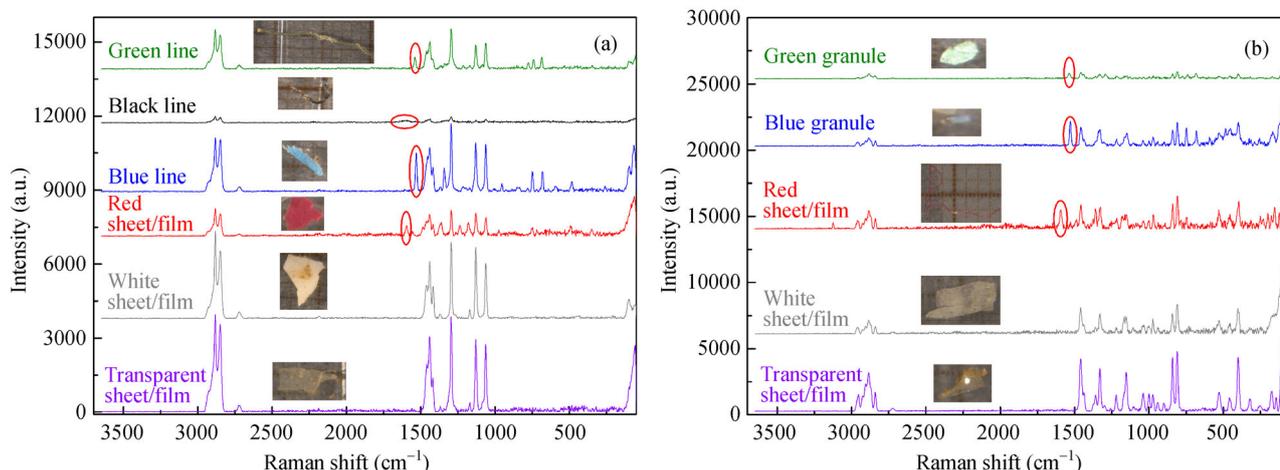


Fig. 3 Raman spectra of field-collected PE (a) and PP (b) MPs with different colors (characteristic peaks of pigments/dyes $> 1000 \text{ cm}^{-1}$ are marked with red rings). The baselines of these spectra were corrected to remove the fluorescence background.

to be from agricultural films in some coastal areas (Tang et al., 2018a). Combined with the fact that sheet/film accounts for the main part of floating MPs (Section 3.3), it is likely that household/agricultural products with low recovery value are the main sources of MPs in this sea area. Household products with low recovery value were one of the main sources found along China's coast (Chen et al., 2019); they might also be important sources for other areas around the SCS.

The H' value of floating MPs in the study area varied from 0.64 to 1.51, with a median of 1.01. The J' values of floating MPs ranged from 0.46 to 0.92, with a median of 0.65. Both the median values of H' and J' were slightly smaller than the result of the mid-west Pacific (1.19 and 0.75, respectively; Wang et al., 2020), but the differences were not significant ($p = 0.136/0.141$ with post hoc LSD, and $p = 0.409/0.422$ with post hoc Bonferroni). In contrary, the median H' value (0.59) in the North-western Pacific (Pan et al., 2019) showed a significant difference from this study ($p = 0.000$ with both post hoc LSD and post hoc Bonferroni), while the difference in evenness (the median J' = 0.73 for the North-western Pacific) was not significant. This result might be because the samples were collected in the 2017 survey in the North-west Pacific Ocean using μ -Raman for identification. Instead, an identification strategy combining μ -Raman with ATR-FTIR applied later enriched our MP database (Table S3 and Fig. S1). It seems that the evaluation of MP diversity is greatly influenced by the identification method and statistical caliber. In this study, the median H' values of floating MPs around Meiji Reef, Chigua Reef and Huayang Reef were 1.08, 0.86 and 0.89, respectively, and the median J' values around these three reefs were 0.68, 0.76 and 0.55, respectively. The differences were not significant ($p > 0.05$).

As shown in Fig. 2 and Table S5, PE, PP and PET were the main components of MPs in the biological samples. According to Wang et al. (2019), PET and PAN (polyacrylonitrile) fibers with diameters \leq approximately 20 μm could be viewed as mostly fabric fibers; additionally, transparent and translucent PE and PP lines with diameters $>$ approximately 20 μm might be from ropes/line/net (mainly used in fisheries). Different from the results in Fujian Province of China in our previous studies (Fang et al., 2019), 58.3% and 37.5% of the fibers/lines found in the southern SCS were attributed to the former and the latter, respectively. This result means that the fiber/line-shaped MPs exposed to local marine life come from both fabric fibers and fisheries/aquaculture activities. The decrease in fabric fibers might be due to the long distance from shorelines. The main sources of fabric fibers are related to residential areas and corresponding human activities such as washing clothes (Browne et al., 2011). It could be noted that the material densities of both fabric fibers (e.g., PET and PAN) were higher than that of seawater (Table S3). Therefore, during the migration of

MPs to the open sea, the ratios of high-density compositions decrease gradually in seawater, leading to their reduction in organisms living in remote sea areas.

4 Conclusions

As a preliminary conclusion, it was found that there was a high abundance of floating MPs in the study area. The diversity based on the composition of MPs was similar to that calculated from a previous survey in the mid-west Pacific, revealing multiple sources of MPs. From the stimulated surface currents, the land-based sources around the southern SCS could be one potential source. The difference in MP characteristics (such as shape) implies that the main MP sources in this area might be different from those in the center of the Pacific Ocean. Household/agricultural products with low recovery value could be suspected sources of floating MPs in this area.

Although the MP abundance in the GIT of fish in this study was basically on the order of magnitude of that found in inshore organisms, the proportion of fabric fibers was significantly reduced; both fabric fibers and fishing/aquaculture activities should be the main sources of MPs exposed to organisms in the open sea.

The high abundance and diversity of floating MPs and their multiple possible sources show that reducing plastic pollution is an issue that needs joint efforts of economies around the SCS. To date, China, Indonesia, Malaysia, the Philippines and Vietnam have restricted or banned the use of plastic bags in all or part of their jurisdictions. Recently, the government of China claimed an ambitious plan to gradually ban the use of nonbiodegradable plastic bags, disposable plastic products for hotels, express plastic packaging, etc., by the end of 2025. These products were abundant in the waste streams of South-east Asia and China, and some of them were also the most likely sources of floating MPs in the SCS in this study. The development of single-use plastic alternative products is a possible trend (Tan et al., 2021; Xu and Ren, 2021). In addition, based on the results concerning MPs ingested by organisms, how to address the improper use/abandonment of plastics in fishery/aquaculture activities might be one of the keys to reducing the risk of MPs in this sea area.

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