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Lost but can't be neglected: Huge quantities of small microplastics hide in the South China Sea

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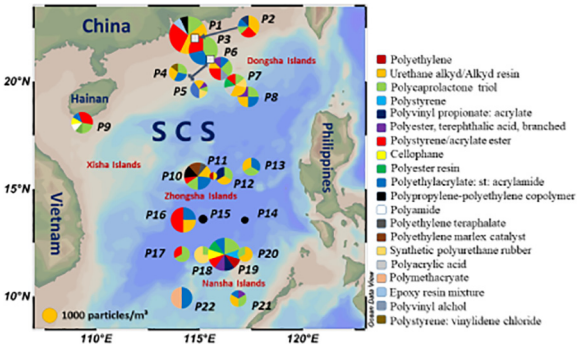
HIGHLIGHTS

- Large quantities of MPs (0.02-0.3mm) were found in the surface water of South China Sea.
- Large-scale distribution characteristics of MPs in a marginal sea were presented, with topographical inventory estimation.
- The distribution pattern of MPs in this study presented a strong terrestrial impact.
- Improvement of the method for smaller MPs are imperative.

GRAPHICAL ABSTRACT



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ABSTRACT

Large quantities of microplastics with small particle sizes were found in the South China Sea (SCS). The abundances of microplastics in seawater were 0.045 ± 0.093and 2569 ± 1770 particles/m^3 according to the bongo net and pumping sampling methods, respectively. Smaller-size fractions (size < 0.3 mm) contributed 92% of the number of microplastics to the total load. Continental slope is the largest reservoir of microplastics with an inventory of 295 tons. 21 polymer types were found in the samples using the micro Fourier Transform Infrared Spectroscopy (FTIR), among which alkyds (22.5%) and polycaprolactone (PCL) (20.9%) accounted for almost half of the total polymer content. Lighter plastics would not only concentrate upon the coastal area, being more likely to drift further into open seas with ocean currents. The distribution characteristics showed that it was mainly controlled by terrestrial input of the Pearl River. This study, as the first report from SCS on microplastics in water for its distribution and influence factors, provided impetus for further research on the transportation fate and the behavior of this emerging pollutant from coastal zone to the open oceans.

1. Introduction

Due to their convenience and economy, global production of plastic has increased from 1.7 to 299 million tons during the last 60 years, and

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has posed serious threats to the ecosystem (Reisser et al., 2015; Plastics Europe, 2015). Transported by wastewater effluent, runoff, rivers and aerosol deposition (Kooi et al., 2016), oceans become one of the significant sinks of waste plastics debris (Andrady, 2017). The annual input of plastic debris into the ocean is increasing rapidly (from 4.8 to 12.7 million), and there might be more plastics in oceans than fish by the year 2050 (Jambeck et al., 2015; Floyd, 2016).

Microplastics are defined as the plastics smaller than 5.0 mm (Thompson et al., 2004). In the ocean, these smaller plastics may derive from environmental aging like UV light, weathering, corrosion and degraded into microplastics. Also, industrial manufacture is a considerable source of microplastics, such as exfoliating facial scrubs, toothpastes and resin pellets (Farrell and Nelson, 2012; Sundt et al., 2014; Browne et al., 2011). With the toxicity and affinity of other hazardous pollutants, which were added during manufacture or persistent organic toxicants absorbed from surrounding water, worldwide microplastics have drawn increasing concern. After ingestion, microplastics can act as vectors by transferring toxic chemicals to the ingested organisms such as invertebrates, fish, birds and mammals, and probably have adverse physical or chemical effects and the health risk (Teuten et al., 2007; Cole et al., 2011; Wright et al., 2013; McCormick et al., 2014; Desforges et al., 2014).

It has been observed that microplastics existing worldwide in beaches, waters and sediments in the shorelines, marginal seas as well as the open oceans (Thompson et al., 2004; Goldstein, 2012; Cózar et al., 2014; Eriksen et al., 2014; Reisser et al., 2015; Lusher et al., 2015; Waller et al., 2017). There are still a number of unanswered questions about microplastics in the ocean, including the most straightforward and fundamental question about the quantity of microplastics (Hardesty et al., 2017). There are some additional key issues need to understand as well, such as the sources, transport and fate. Many of the studies focused on quantifying the contamination levels and considered their potential effects on biota and the environment (Thompson, 2016), but were lack of quantitative and systematic studies on the influence factors of its distribution (Browne et al., 2011; Ivar do Sul et al., 2013). From the coastal to the open oceans, marginal seas act as important media in contaminants transportation, therefore studies on microplastics in the marginal seas were needed.

The South China Sea (SCS) is the largest semi-enclosed marginal sea located in the western Pacific Ocean, with a seasonal monsoon climate (Cai et al., 2017). The SCS has unique, complex hydrological conditions, including energetic circulations such as the basin-wide, sub-basin and mesoscale gyres (Li, 2002). The way of currents and winds can facilitate the nutrients supply and food web transfer of toxic chemicals as well (Liu et al., 2002). The SCS provides a habitat with abundant planktonic life for a diverse array of benthic, mid-water species and top predators (Sun et al., 2017). Also, the northern SCS is an important fishery ground (Wang et al., 2015), it contains lucrative fisheries that are crucial for the food security of millions in Southeast Asia (Barber, 1998). On the other hand, the SCS is under considerable anthropogenic pressure, surrounded by several developing countries with large population, intense industry, agriculture and shipping, coastal cities, ports, shipping activities, uncontrolled coastal landfills and dumping sites along the coast are also the important source of pollutions (Jambeck et al., 2015).

In this research, seawater samples were collected from the surface to 200 meter depth in the SCS in the April of 2017 with two methods. This study aimed to (1) present, for the first time, the abundance, compositions and sizes of microplastics in the SCS; (2) estimate the inventory of microplastics and potential ecological risk in the SCS; (3) investigate the distribution characteristics, possible source and the anthropogenic activities factors may impact on their distribution; (4) explore the influence of surface current on the transportation of microplastics.

2. Materials and methods

2.1. Field sampling

2.1.1. Study area

Sampling was conducted aboard the central SCS oceanographic research cruises (NORC-06) from March to May 2017 by the R/V

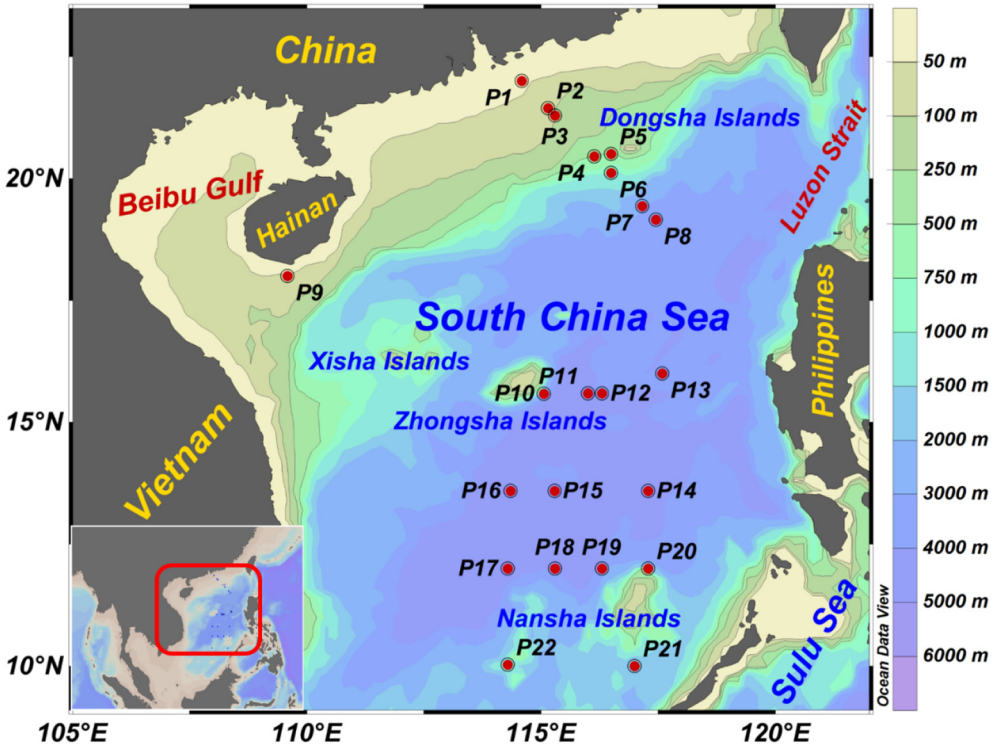

Fig. 1. Sample stations (P1–P8 are located at the Pearl River section, P9 is close to the Hainan province, P10–P22 are located at sea basin in the central of the SCS. The depth contours are 50, 100, 200 and 500 m).

ShiYan I. The SCS is the largest marginal sea in the Southeast Asia and join the Pacific Ocean through the Luzon Strait, which has with a sill depth of about 2400 m (Cai et al., 2016). Other important passages include the Taiwan Strait (connect to East China Sea), Mindoro Strait and Balabac Strait (connect to Sulu Sea), Karimata Strait (connect to Java Sea) and Malacca Strait (connect to Andaman Sea) (Huang et al., 2016). It is bounded by the Asian continent, Philippine Archipelago, and Great Sunda Islands, the Pearl River Estuary is the largest river system flowing into SCS and covers an area of >8000 km^2 during periods of peak discharge with a watershed of 230,000 km^2. The SCS is affected strongly by monsoon, thus its surface circulation is mainly driven by the monsoon and has significant seasonal vibration. In winter the SCS is dominated by the strong northeasterly monsoon, whereas in summer the winds reverse direction to southwesterly. As a result, there is a large cyclonic gyre in the SCS in winter, while a weak cyclonic gyre remains in the northern SCS in summer, but an anti-cyclonic gyre occupies the southern SCS (Liu et al., 2008). In spring, a general cyclonic gyre is still maintained in the SCS but is weaker than that in the winter. Considered to get samples in different size class and getting enough particles for analysis from the relatively clean sea water in the SCS, two pervasive sampling strategies: trawl of bongo net and pumping water (Fig. 1) were designed in this survey.

2.1.2. Sampling methods

The 200 m water column samples were collected by the bongo sampler, connected to a central axle and a pair of 333 μm mesh nylon plankton nets in the 19 stations (including another two duplicated samples in one site). Totally 110 m^3water from 19 stations was filtered by this method at the cruise. The bongo trawl was lowered to 200 m depth by paying out 218 m of wire at 1 m/s, then the net was retrieved at 0.5 m/s (Doyle et al., 2011). Contents of the net were washed into a pre-cleaned glass jar with Milli-Q water, and fixed in 30% formalin for further processing in laboratory.

To detect the smaller particles, the surface water was taken by a pump and filtered at 22 stations. It was a centrifugal pump made of titanium and stainless-steel progressive connected with double water meters and PVC water hose (Zhao et al., 2014). The seawater intake pump was located at 0.5 m below waterline on the quarterdeck bow side of the vessel for approximate 3 m^3. The flux was recorded by double water meters, also estimated according to the sampling time. All the samples were pass through three square filter meshes with 5 mm, 154 μm and 44 μm, respectively. The substances remain on the 5 mm mesh was discarded. The meshes with 154 μm and 44 μm were enwrapped with aluminum foil and stored at 4 °C until analysis. In order to avoid contamination, samples were collected when the vessel stopped for standardly hydrological cruise sampling stations (Fig. 2).

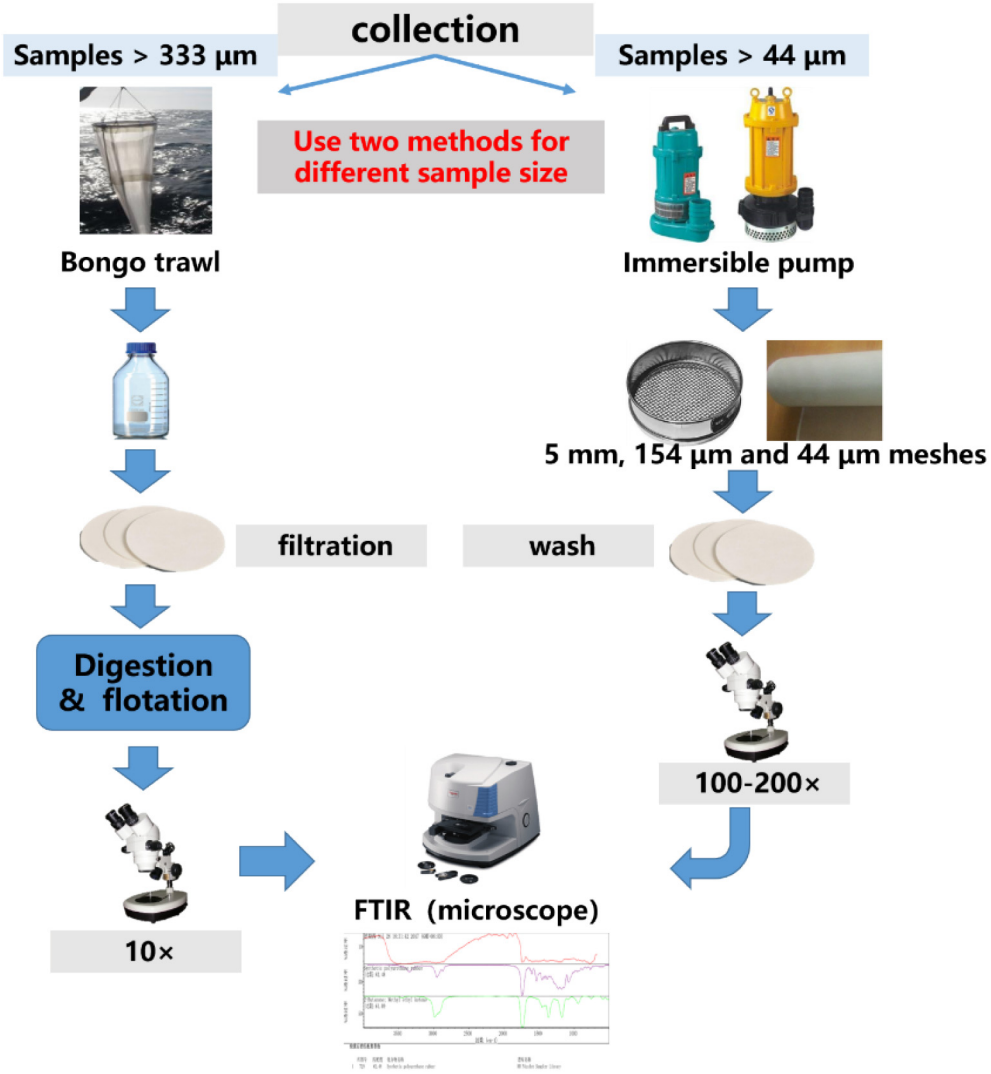

Fig. 2. Sample collection and treatment.

2.2. Sampling treatment

2.2.1. Digestion and flotation of trawl samples

The samples from trawl were disposed with digestion and flotation. Firstly, pre-filtered through a 5 mm metallic mesh and large debris (>5 mm) in the water samples then discarded. The rest samples were filtered under vacuum onto nylon filters (Millipore, 20 μm), and the contents on the filters were oxidatively cleaned using 30% H_2O_2 (Nuelle et al., 2014; Zhao et al., 2014). After filtering the digestion solution with nylon filters (20 μm), plastic particles were separated via flotation in a NaCl solution (1.20 g/cm^3) for 24 h (Nuelle et al., 2014), and then the supernatant was collected onto nylon filter following filtration of the saline mixture.

2.2.2. Ultrasonic cleaning of mesh samples

The pumping samples on meshes were washed by 200–300 mL Milli-Q water with ultrasonic cleaning instrument 5 min for three times. Then the washing solutions were mixed together and pass through the nylon filters (Millipore, 20 μm) under vacuum. After washing down, we put the filters and meshes under the microscope to watch if there is any residue.

The samples on meshes were disposed with a non-invasive and non-destructive methodological approach, because it maintains the primitive amount, size, and form of microplastics for investigating. In fact, methods such as density separation (using salt solutions), or digestion by chemicals (acids, alkalis, enzymes, and solvents) are either too expensive, time-consuming, or significantly affected the samples themselves (Hidalgo-Ruz et al., 2012; Claessens et al., 2013; Cincinelli et al., 2017). Indeed, it has been found that the treatment with 30% H_2O_2 would alter the characteristics of the polymer, by making it difficult to identify or in some cases even dissolving it, particularly with regard to fibers (Crawford and Quinn, 2017; Claessens et al., 2011). Thus, in our research this purification step should only be undertaken if there is a high degree of organic matter present on the samples which is hindering visual selection of the microplastics (e.g. samples by trawl in this research).

To the best of our knowledge, this is the first survey of the abundance, composition and distribution of microplastics on the SCS surface water, also one of the few studies using mesh of 44 μm to ensure that most of the micro-debris were remain on it with little loss. Thus, we select analyze without any potentially invasive pre-treatment of the samples on meshes. We hope to investigate the filters after the first filtration directly without any other pre-treatment (except drying) that might remove the particles on the filters' surface. That's likelier to be the true level of real microplastic samples on field.

2.3. Identification and analysis

The nylon filters with larger samples by trawl were put under fluorescence microscope (Leica, M165FC, Switzerland) at 10× magnification and taken photos using “zigzag” pattern until making sure that everywhere had been covered.

The nylon filters of pumping samples were put under a microscope (ZEISS, Scope A1, Germany) at up to 100–200× magnification. Due to the high magnification, we have to shoot at least 2000 photographs to cover one nylon filter. Considering the time and resources required, we prefer to shoot 200 photographs at each station at random, and cover one station with 1000 photographs for statistical estimate of

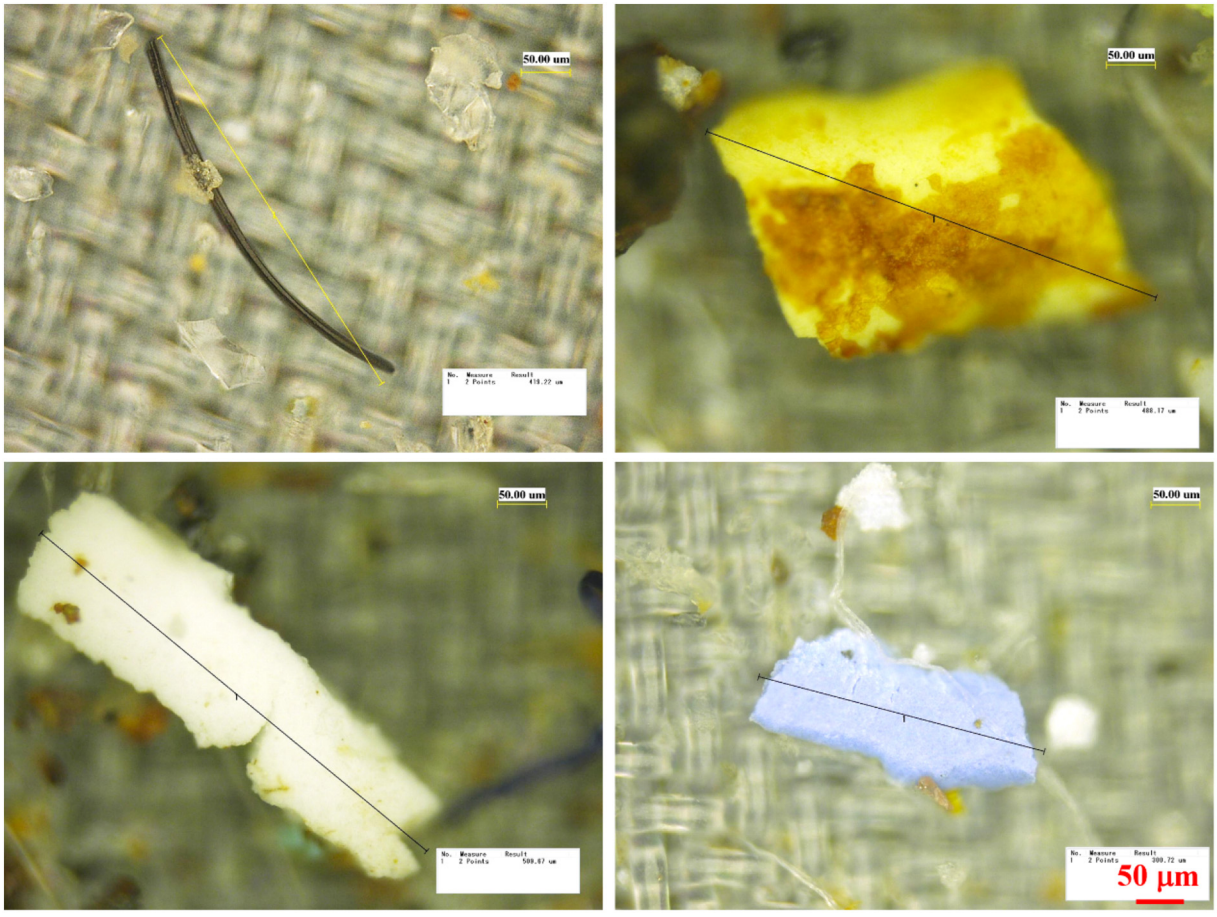

Fig. 3. The abundance of microplastics (detected particles from 44 μm mesh with size of 419.22 μm, 488.17 μm, 509.67 μm and300.72 μm), in each sample showed an evident difference between the microplastics found in the samples collected by trawl net (Fig. 4).

error. Then careful visual sorting, necessary of the counting process of microplastics, was conducted (Hidalgo-Ruz et al., 2012). To avoid misidentification and underestimation of the plastics from other materials, such as organic debris (shell fragments, dried algae, or seagrasses, etc.), this study follows the criteria implemented by predecessors' research (Hidalgo-Ruz et al., 2012; Crawford and Quinn, 2017).

Since the available and veracity of the Fourier Transform Infrared Spectrometer (FTIR) which could be used for the identification of small debris diameter, we selected particles each station randomly to test the polymer types using a FTIR (Thermo Fisher, Nicolet 6,700, U.S.A.) for large size microplastics (0.3–5 mm) and a Micro Fourier Transform Infrared Imaging microscope (Thermo Fisher, Nicolet iN 10 MX, U.S.A.) for small size microplastics (0.02–0.3 mm) (Tagg et al., 2015; Vianello et al., 2013). When microplastics were identified by micro-FTIR, each piece of microplastics was detected by at least two different sections (Martins and Sobral, 2011) and the highest similarity (at least 60% similarity for confirmation) in the database were assigned to the microplastic samples. The spectra that we got were processed by MONICTM PictammmmTM software and compared with the OMNIC polymer spectra library (Mecozzi et al., 2016).

Ocean Data View 4.5.6 was used for Figs. 1 and 5. Figs. 3, 4 were taken by microscope imaging system of Keyence VHX-600E. Size distribution was calculated by Origin 2018. Type distribution and inventory calculation were performed using Microsoft Excel.

2.4. Quality control and quality assurance

The sampling device and all filter meshes were thoroughly cleaned with deionized water before use. At sampling stations, the meshes were removed from the filter cartridge, immediately folded, wrapped with aluminum foil, transferred to a compact bag and sealed. Three field parallel samples of trawl net and two laboratory blanks were conducted.

When laboratory treatment and microscopic examination were conducted, strict control on the possible air contamination is proceeded as well. First of all, we hold a dress code of non-plastic material (lab coat, head cover, nitrile gloves) and cleaned laboratory platform, washed hands and forearms were measures to prevent contamination from adhering dirt particles. Prior to use, all instruments and vials were cleaned with deionized water. All openings of sampling and analysis devices (e.g. filter unit, funnels in the vacuum pump) were covered with aluminum foil when not in use and placed in direct proximity to the work area, providing a control of potential airborne contamination (Lenz et al., 2015). After the microscopic examination, we would put the lid on immediately (Nuelle et al., 2014).

Secondly, we conducted double mesh laboratory blanks during the whole treatment process, in which 250 mL of Milli-Q water stored within sample containers were treated the same as field samples. These two laboratory blanks were taken examination under microscopic, no plastic was found on the filters, implying that contamination from the containers, lab or processing was negligible.

3. Result and discussion

3.1. Abundance of microplastics in the SCS

3.1.1. Larger microplastics (0.3–5 mm)

Average concentration of larger particles (size > 333 μm) in the SCS was 0.045 ± 0.093 particles/m^3, and this result was comparable with other studies in which the net trawls with 0.3 mm mesh were

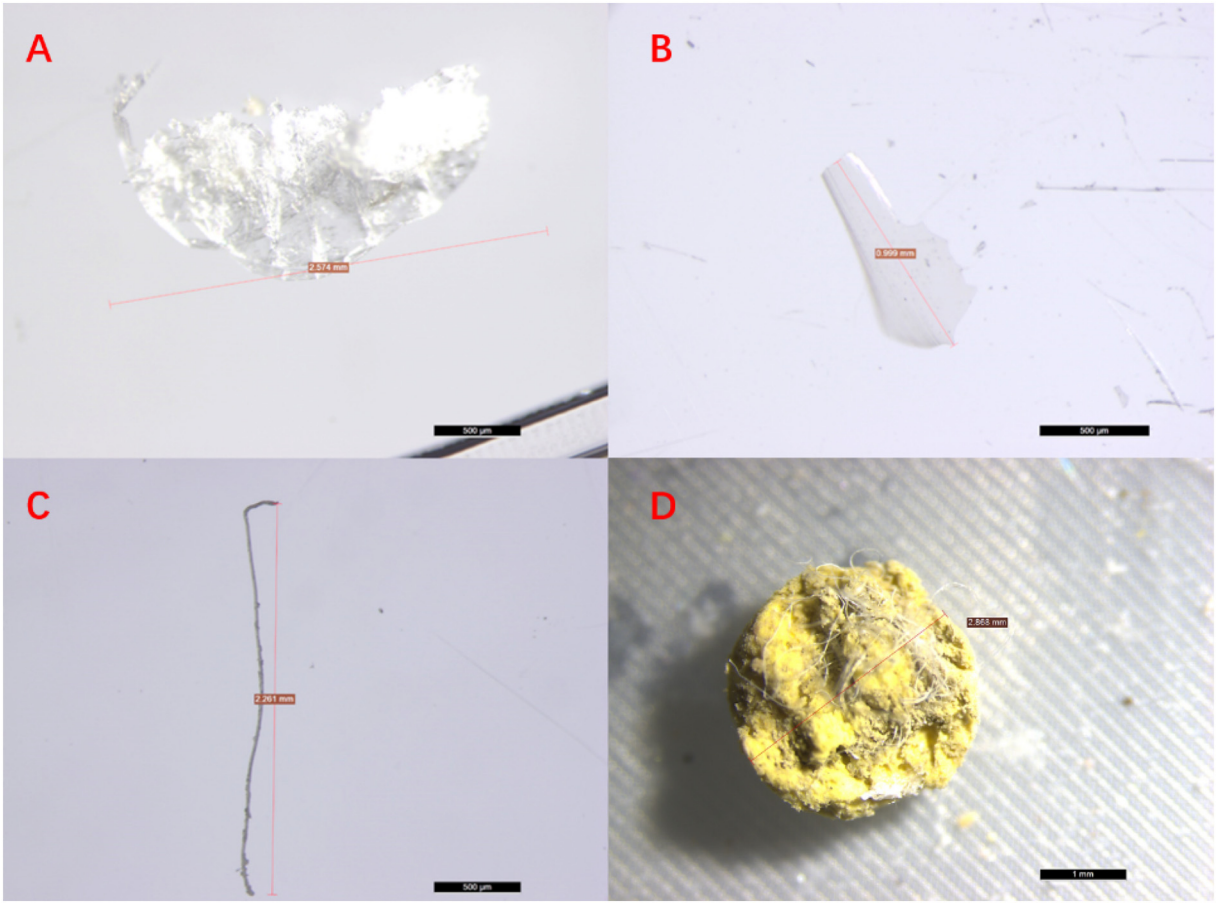

Fig. 4. Particles selected from trawl samples. A and B showed plastic particles, C and D showed the particles with natural or other composition. A: Polypropylene-polyethylene copolymer with size of 2.574 mm, B: Polyethylene: propylene with size of 0.999 mm; C: Menthol acetate with size of 2.261 mm, and D: Raw material for ceramics with size of 1.400 mm.

used (Table S1 in SI). Compared with other results, the concentrations founded here were in the same order of magnitude of those in Bohai Sea (0.33 ± 0.34 particles/m^3) and the East China Sea (0.167 ± 0.138 particles/m^3) (Zhao et al., 2014; Zhang et al., 2017). Higher concentrations were found in the coastal areas near the Korea (47 ± 192 particles/m^3) and Japan (1.72 pieces/m^2) (Song et al., 2014; Isobe, 2016), indicating the importance of terrestrial input near the industrialized and populated coastal areas (Browne et al., 2011; Cheung et al., 2016). Besides, the difference of sampling methods may give rise to the concentration variations. Unlike the Manta trawl collecting the surface water only, where plastics are more likely to exist, results from vertical trawling would be smaller due to the dilution.

We gave appropriate conversion to the reports in different units for better understanding and comparison (Table S1 in SI). Generally, the abundance of microplastics in seawater was in the same order for marginal seas or open oceans, such as north western Mediterranean Sea (0.116 particles/m^2), Sardinia Sea (0.13 ± 0.27 particles/m^3), southeast Bering Sea (0.045 particles/m^3), Tasman Sea (2174 ± 1071 pieces/km^2), Ligurian Sea (0.94 ± 2.55 particles/m^3), and Northeast Atlantic Ocean (2.46 ± 2.43 particles/m^3) (Doyle et al., 2011; Collignon et al., 2012; Lusher et al., 2014; Rudduck et al., 2017). However, our results are nearly three orders of magnitude lower than those sampled using mesh size <333 μm, which indicating the significance of the samples with smaller particle size.

3.1.2. Smaller microplastics (0.02–0.3 mm)

Microplastics were detected at all stations, but the concentrations varied considerably ranged from 300 to 7467 particles/m^3, with mean value 2569 ± 1770 particles/m^3 (Fig. 3).

The lack of the microplastic data of whose particle sizes were smaller than 333 μm, prevents the comparative analysis with many of the previous studies. Microplastic concentrations reported here are in the same order of magnitude as in the studies sampled using mesh size <333 μm whether sampled by net, pump or bulk water, in the black sea (1100 ± 900 particles/m^3), coastal waters of Sweden (150–2400 particles/m^3), NE Pacific (2080 ± 2190 particles/m^3), Yangtze Estuary, China (4137.3 ± 2461.5 particles/m^3) and in the SE Korea coastal waters (1143 ± 3353 particles/m^3) (Noren, 2007; Desforges et al., 2014; Zhao et al., 2014; Aytan et al., 2016) (Table 1).

It has been found that five natural zooplankton groups in the northern SCS did ingest microplastics, major of which were fibers and the average length was 167 μm (Sun et al., 2017). The encounter rates of microplastics/zooplankton increased with trophic levels, suggesting that ingested particles can be transferred and biomagnified (Desforges et al., 2015; Erni-Cassola et al., 2017). It is properly that get the one order of magnitudes lager microplastics in the water.

Since there is limited universally recognized protocols for collection, sampling methods vary considerably among researchers, and this will have a strong influence on the ability to detect microplastics as well as the types, sizes, and abundance of microplastics recorded. Anyway, this research still proofs the domain particles in the open areas are in small size smaller than 0.3 mm. The prevalent methods of trawl are not appropriate for the sampling in the open oceans since it will lose most of the microplastics in the area.

Table 1
Microplastic concentrations of surface seawater from each stations.

Station	Sample method	Longitude	Latitude	C (particle/m^3)	Depth (m)	Wind (m/s)
P1	Pump	114.59	22.00	7467	63	7.30
P2	Pump	115.15	21.44	3810	96	9.10
P3	Pump	115.30	21.29	4375	114	8.40
P4	Pump	116.14	20.45	2745	438	3.80
P5	Pump	116.50	20.50	2778	698	3.40
P6	Pump	116.50	20.11	2963	817	1.40
P7	Pump	117.16	19.43	3889	3021	2.00
P8	Pump	117.45	19.15	2222	3673	4.30
P9	Pump	109.59	18.00	2308	98	9.30
P10	Pump	115.06	15.58	3768	3466	6.20
P11	Pump	116.00	15.59	360	4189	2.90
P12	Pump	116.30	15.59	1429	4153	4.30
P13	Pump	117.59	16.00	1667	4050	1.70
P14	Pump	117.29	13.59	301	4179	4.90
P15	Pump	115.29	13.59	444	1470	4.00
P16	Pump	114.35	13.59	3265	3504	2.70
P17	Pump	114.29	12.00	1333	29 56	9.00
P18	Pump	115.30	12.00	1481	4378	6.60
P19	Pump	116.30	12.00	5556	3001	4.70
P20	Pump	117.29	12.00	952	2958	8.60
P21	Pump	117.00	10.00	1190	1639	6.70
P22	Pump	114.29	10.03	2222	2042	3.60
T1	Bongo net	115.50	12.00	0.00	4194	11.4
T2	Bongo net	117.50	12.00	0.00	3330	4.2
T3	Bongo net	117.00	12.10	0.17	2604	9.2
T4	Bongo net	117.99	12.98	0.00	3921	6
T5	Bongo net	117.50	13.00	0.00	4143	7.6
T6	Bongo net	115.50	13.00	0.00	1470	6.5
T7	Bongo net	114.50	10.07	0.00	1284	9.4
T8	Bongo net	116.50	10.00	0.17	1648	6.1
T9	Bongo net	117.50	10.00	0.00	1639	10.1
T10	Bongo net	117.00	14.00	0.00	4233	6
T11	Bongo net	110.50	18.00	0.00	159.7	9.7
T12	Bongo net	112.00	18.00	0.33	2479	4.2
T13	Bongo net	112.50	18.00	0.00	2119	4.4
T14	Bongo net	114.50	18.00	0.00	2089	1.2
T15	Bongo net	115.50	18.00	0.00	3935	3
T16	Bongo net	118.00	16.00	0.17	4050	4.5
T17	Bongo net	117.50	16.00	0.00	4123	2.7
T18	Bongo net	118.00	13.99	0.00	4188	7
T19	Bongo net	116.50	12.00	0.00	4200	1.6

3.2. Composition of microplastics in the SCS

3.2.1. Large microplastics (0.3–5 mm)

Five particles were identified as plastics, as the 3 components of polyester, polyethylene and polypropylene-polyethylene copolymers. They are the most widely used polymer plastics in our daily life and have a large range of products (Hidalgo-Ruz et al., 2012), including packaging, textiles and fishing gears. As expected, among the samples which were taken by smaller pore-size net, we still found these plastic components contributed a lot to the total plastic composition. This is part of the evidence that macroplastics are the significant source of microplastics. As the plastic debris with a wide range of sizes enters the marine environment, breakdown process, such as ultraviolet irradiation, high temperatures, and mechanical shear forces, finally degraded plastics into smaller particles even with size <100 μm (Erni-Cassola et al., 2017).

3.2.2. Small microplastics (0.02–0.3 mm)

21 component types were found in the samples using the micro FTIR spectral analysis (Fig. 5). The abundant components were alkyd resin (22.5%), polycaprolactone (PCL) (20.9%), and poly ethyl acrylate (PEA) (15.5%), followed by polystyrene (PS) (14.7%), synthetic polyurethane

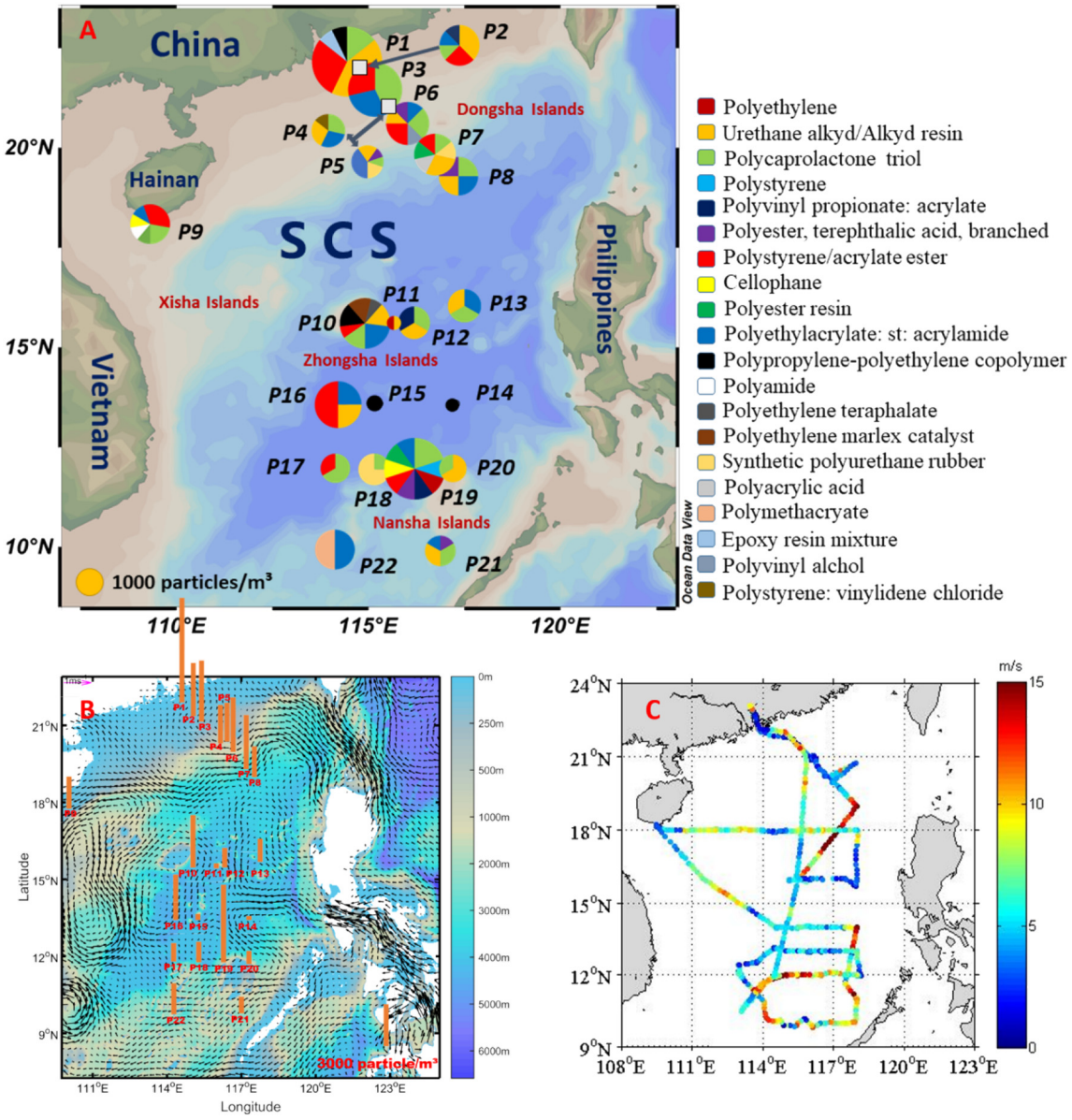

Fig. 5. A: The concentration and component distribution of microplastics in the SCS (totally 21 polymer types marked in different colors, the area of the circle represents the large of concentration). B: The average of surface current in the SCS in the sampling period from March 2017 to April 2017 (the current was derived from the GEKCO (Geostrophic and EKman Current Observatory) product). C: Beaufort wind scale of sampling area (source: Real-Time Collector on the R/V ShiYan I)

rubber (PTFE) (4.7%), polypropylene-polyethylene copolymer (3.8%), polyester terephthalic acid (3.8%) and other polymers (14%).

Alkyd resin, as the most common component, is a polyester modified by the addition of fatty acids and other components (Wang et al., 2000) and always becomes the raw material of industrial painting including marine application such like anticorrosive purposes on metal or wood on ships (Song et al., 2014). One-third of the world's shipping passes through the SCS carrying over three trillion U.S. dollars in trade each year (Morton and Blackmore, 2001). Originated from paints and the fiber-reinforced plastic (FRP) matrix used on ships, may cause the high level of alkyds (Song et al., 2014). Moreover, another frequently present component is polycaprolactone, which was usually used in the medical instruments and adhesive. The potential environmental impact arising from trade activities, such as shipping for commercial and scientific purposes, and from the sewage treatment plant, was also evidenced.

Polypropylene (PP), polyethylene (PE), styrene acrylonitrile (SAN), polystyrene (PS), nylon, polyvinyl alcohol (PVA) and acrylonitrile butadiene styrene (ABS) are commonly present in both seawater and sediment in the Asian marine environment (Ng and Obbard, 2006; Heo et al., 2013; Noik and Tuah, 2015; Fauziah et al., 2015; Fok and Cheung, 2015; Kim et al., 2015; Tsang et al., 2017). In the continental shelf and slope, these polymers are still present. Polypropylene-polyethylene copolymer was also firstly appeared in deep basin areas. Polymers with low density (<1.20 g/cm^3) are more likely to transport into far-reaching areas in the ocean with waves.

3.3. Spatial characteristics of microplastics and it influence factors (0.02–0.3 mm)

3.3.1. Northern SCS

The highest microplastic concentration was found at site P1 (7467 particles/m^3), which is a site influenced by a mix of waters coming from four rivers' discharge into the SCS, for instance Pearl River and Han River. The lowest microplastic concentration was found at site P14 (300 particles/m^3), at the central of the SCS, with no other input

source. The concentration at sites nearshore (P1) was 25 times greater than sites offshore (P14) in the SCS, indicating a significant riverine contribution and the transportation by coastal currents. Apparently, the microplastic abundance showed a gradual decreasing trend along the section from the Pearl River Estuary into the central of the SCS (23–20°N). This section was located near the Guangdong province which is populated and with large plastic production province in China. In 2015, 9.76 million tons of plastics were produced in the Guangdong province. Synthetic fibers with polyester, polyamide, acrylic and polyolefin are being high related to the population of the area (Salvador et al., 2017). Such high concentration regarded as the large population and economic development.

However, along the shelf break (stations P4 and P5), part of subsurface water reaches the surface as a result of upwelling. Thus, the waters contain a mix of waters originating in different parts, and the microplastics concentration of the surface seawater in station P4 and P5 was relatively low (2745 particles in P4 and 2778 particles in P5).

The section of continental shelf and slope shows that not only the concentration but also the component diversity decreased gradually from the coastal area to the abyssal basin of the SCS, and reduced to the minimum in the P8 with only 4 types of polymers. The coastal stations (P1–P3) all included the component of PS, it is one of the most produced polymers worldwide which could apply to foam food container and packaging. The study on 8 beaches from Guangdong province shows PS foams and fragments being more abundant in the smaller size classes (Fok et al., 2017). This is consistent with our research in polymer variety, which proofed that microplastics originated from land source output in somehow. Sites P8 contained the fewest types of plastic components, with the density from 0.98 to 1.20 g/cm^3, which are the low-density plastics and more likely to flow with ocean currents.

Four types of plastics were identified in sediments from the Beibu Gulf and the coastline of China Sea, namely polyesters, polyethylene terephthalate (PET), polyethylene (PE) and polystyrene (PS) (Qiu et al., 2015), which are in consistent with results near the Hainan province. This consistency is demonstrated the deposition of microplastics (Frère et al., 2017). In this area, polyamide is the first, and, the only, appeared in our samples, Hainan province is the one of the largest provinces of plastic production, the synthetic fiber polymers production reach to the 2 million tons. High concentration is related to this high produce.

Oceanic floating materials including plastics will be transported to a place where is far away from its source area (Brunner et al., 2015). Thus, the plastic particles might converge in a place where local circulation acts to retain such plastics. It seems that the shelf region in the NSCS has a high concentration fishing aquaculture installation, which might be a potential local source for plastics (e.g. Hinojosa and Thiel, 2009) Meanwhile, several harbors and recreational fishing activities probably contribute to the microplastics pollution to the sea. Here, the surface current plays an important role on the plastic debris distribution and abundance in the SCS especially in the NSCS. In a word, whatever the dominant source of microplastics in the SCS, it seems that the SCS represents a vulnerable receiving environment which is sensitive to the pollution.

3.3.2. Central and southern SCS

The average concentration the central of the SCS (between 17°N and 10°N) is 1840 particles/m^3, which is less than half of the nearshore level. Owing to the geological and topographical conditions, the seawater in the central SCS has weak exchange with land and thus maintains low accumulation. Two distinct high values in the central of the SCS appeared on the sites near the Xisha Islands (P10) and Nansha Islands (P19 (Fig. 5)). This provided an evidence that the microplastics have local source.

A recent study showed that atmospheric fall out is also a considerable terrestrial source of synthetic fibers thus the southerly winds may have also increased wind transport of fibers from land to sea (Dris et al., 2016). In this study, we estimated fitted curves between concentration for such stations and the wind speed at the 95% confidence intervals (CIs) (Fig. 4 in SI). There was no clear pattern of higher microplastic concentration in inshore stations than in offshore stations (Fig. 5). Also, no clear relationship was found between air transport by wind flow and microplastic spatial distribution. This suggests that other processes besides the large-scale circulation field would not govern microplastic spatial distribution.

In the central of the SCS, stations P10 (3768 particles/m^3) and P19 (5556 particles/m^3) are the two stations with the maximums of concentration and component diversity. In SCS waters, microplastics may be discharged in wastewater from islands, coastal areas of the Southeast Asia and China also from research, fishing and tourist vessels. Station P10 and P19 are located near the Xisha islands and Nansha islands respectively, these islands are the inhabited islands. The daily use of microplastics in personal care products was estimated to be in the range from 2.4 to 27.5 mg per day per person (Gouin et al., 2011, 2015). However, some information is not available, such as the volume of waste water released into the SCS and empirical data on microplastics in the wastewater.

3.4. Microplastic inventory in the SCS

The inventory of microplastics in the SCS was calculated as follows:

$$I_{ix} = C_{ix} \times S_x \times H_x,$$

where I_{ix} is inventory, C_{ix} is average content, H_x is depth of the sea area, we selected as 5 m, S_x is the area of different topographies.

The inventory of microplastics in the entire SCS (continental shelf, continental slope, and deep basin) was estimated to be $4 \pm 3 \times 10^{16}$ particles, equivalent to volume of 600 m^3 and weight of 700 tons respectively. The inventory of microplastics was dominated by their masses, the continental slope is the major compartment with an inventory of 295 tons, whereas the continental shelf and deep basin are estimated to store 156 tons and 234 tons, respectively. Among the compartments, continental slope was the largest reservoir of microplastics.

According to the research on the worldwide 192 countries and regions with live within 50 km from the coastline population emissions of plastic, the top ten over eight largest emitters from Asia, in 2010 they emitted more than half the world's rubbish. The six coastal SCS countries contribute 2.56–7.08 million tons among which China emitted 1.3–3.53 million tons (Jambeck et al., 2015). By reason that plastic waste dribs only a matter of the source of microplastics and these countries have long coastlines (China is surrounded by the Bohai sea, the east China sea and the SCS), rivers ought to flow into varies seas. On the other hand, this inventory merely covers the microplastics float on the surface water, there has been evidence shows deposition process of plastic debris indicated the deep sea as the reservoir of the plastics (Woodall et al., 2014). Thus, this is reasonable that the estimated microplastics inventory in the ocean is one to four orders of magnitude smaller than the reported global emission.

The huge amount of pollutants implies a great ecological risk. Smaller particles in this study are more likely to be ingested by marine organisms and transported through the food chain. So far, no detailed studies of microplastic particles as well as paint particles have been conducted focusing on very small microparticles (1–50 mm), in either marine or terrestrial ecosystems (Imhof et al., 2012). However, it has been reported that the virgin or colored particles contained a high variety of metals such as cadmium, lead and copper, whose potential human exposure pathways have been recognized, especially for those with the size smaller than 50 mm (Imhof et al., 2012; Wright and Kelly, 2017). The upper ocean water provides important living environments for large of the marine biota, and serves as the most productive zone,

where the smaller microplastics would pose a more serious ecological threat.

3.5. Influence factors of microplastics

3.5.1. Influence of methods

This research uses the mesh net with pore size of 44 μm for samples collection. Compared to prevailing sampling methods, net with 333 μm even more lager, this method would render more particles remain on it. There has proven that concentrations in Swedish waters were up to 100,000 times greater when sampled with 80 μm rather than a 450 μm mesh (Noren, 2007). And the comparative methods in the southern coast of Korea show mean abundance of microplastic particles with hand net (50 μm) are 1143 ± 3353 particles/m^3, which is larger than Manta trawl (330 μm) 47 ± 192 particles/m^3 (Song et al., 2015). The results are consistent with us, neuston nets with mesh >333 μm may result in the loss of small particles.

Furthermore, there aren't any pre-treatment processes that might remove the fibers or alter their primitive form except ultrasonic cleaning on the samples in the lab (Cincinelli et al., 2017). On one hand, we intend to get the non-destructive and full-scale samples to acquaint the actual state of the particles in the SCS. On the other hand, when we scan the samples under the microscope, seldom of them present observable organic or cellular structures. Thus, it is reasonable for studies to choose different pre-treatment methods for samples with different degree.

3.5.2. Size distribution of the small particles (size > 20 μm)

As divided particles into three size classes, 8% of them are larger than 0.3 mm, 0.1–0.3 mm count for 43% of all the particles and mainly, more than half of the focus on the range smaller than 100 μm roughly (Fig. 6). The 0–100 μm size fraction was the most abundant of all size classes at our sampling stations. Thus, the smaller particles we found in this study are in high quantity. The size of these particles found in this study ranges from 0.8 mm to 20 μm and intensive distribution in the level smaller than 200 μm. Recently study from sea surface tows shows that a power-law increase in small microplastics (i.e., <1 mm) with a decreasing particle size (Erni-Cassola et al., 2017). Average particle size of microplastics in surface waters increases exponentially with decreasing particle size (Song et al., 2014).

Overall, these results highlight the important contribution of smaller-size fractions to the total plastic load. The results demonstrated that in the SCS, plastics larger than 0.3 mm made limited contribution to the total amount of microplastics. According to the estimation, about 275 million metric tons of microplastics was generated in the coastal countries, with 4.8 to 12.7 million of them entering the ocean (Jambeck et al., 2015). However, the inventory of microplastics in the worldwide open oceans was significantly smaller than their input into the marine environments. Our results therefore may help to resolve the speculation about the "apparent" loss of this fraction from surface waters, the smaller microplastics <0.3 mm which broad spatial extent in the open oceans.

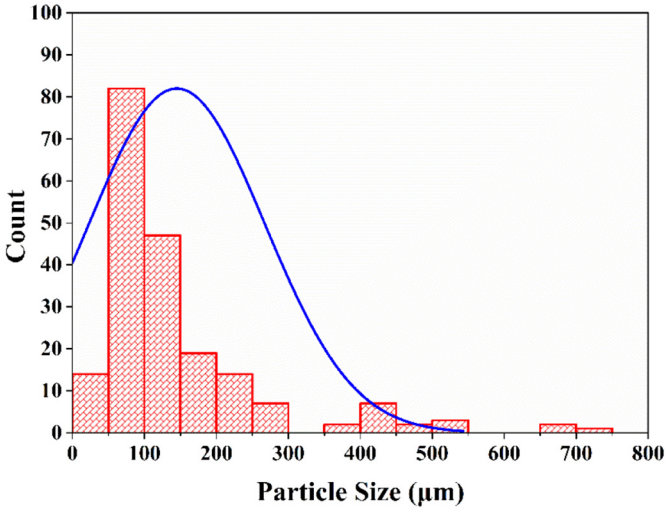

Fig. 6. Particle size distribution in surface water (n = 200, counted as interval of 50 μm, max = 719.07 μm, min = 19.06 μm, the average length is 145 μm, there were 14, 82, 47, 20, 14, 2, 5, 16 particles <0.05 mm, 0.1 mm, 0.15 mm, 0.2 mm, 0.25 mm, 0.3 mm, 0.4 mm and 0.72 mm respectively).

4. Conclusion

Our research highlighted the existence of smaller microplastics (0.02–0.3 mm) in the open SCS, the average particle size of microplastics in surface waters increased exponentially with decreasing particle size. The concentration of particles >333 μm is 0.045 ± 0.093 particles/m^3, with the mean value of smaller particles (0.02–0.3 mm) being 2569 ± 1770 particles/m^3.

This distribution characteristics showed that for the terrestrial pollutants, the Pearl River estuaries act as a gateway to the oceans, while development of the reefs and islets in central and southern part cause the high pollution level. Low-density plastics will not only concentrate upon the coastal area, they are more likely drift further into ocean central by wave action. Continental slope is the main reservoir of microplastics.

This may be especially significant in continental freshwater and terrestrial environments, where concentrations of these particles were expected to be higher than those in the marine systems, due to the proximity to the use of these particles (Dris et al., 2016; Lohmann, 2017). One concern of the impact of microplastics on marine ecosystems is its transfer across marine food web, which may cause negative effects chemically and physically (Teuten et al., 2007). Further research on the transportation fate and the behavior of this emerging pollutant from coastal zone to the open oceans needed to be conducted.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2018.03.197.

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