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Spatiotemporal distribution, source identification and inventory of microplastics in surface sediments from Sanggou Bay, China

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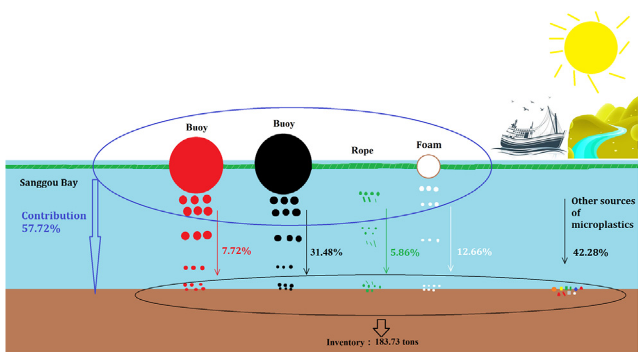
HIGHLIGHTS

- Microplastic pollution in sediments was at a heavy level compared with other areas.
- Approximately 57.72% of microplastics derived from mariculture plastic facilities.
- Estimated microplastics inventory in sediments of Sanggou Bay was 183.73 tons.
- Mariculture is an important contributor to sedimentary microplastics of Sanggou Bay.

GRAPHICAL ABSTRACT



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ABSTRACT

Microplastic pollution has become a major global environmental issue. Yet the source identification and inventory of microplastics in mariculture areas remain unclear. Here we investigated the spatiotemporal distribution, source and inventory of microplastics in surface sediments from Sanggou Bay, China. The results showed that average abundance of microplastics in the surface sediments was 1674 ± 526 items/kg dry weight, which represented a heavy level when compared with other sea areas, including coastal waters, estuaries, the open sea and other mariculture areas. Microplastics with a size of <0.5 mm were dominant throughout four seasons. The dominant shape of microplastics was granule in summer, autumn and winter, and film in spring, respectively. The most common color of microplastics was transparent. Polyethylene was the dominant polymer in summer, autumn, and winter, while polystyrene accounted for the largest proportion in spring. Approximately 57.72% of the microplastics in surface sediments originated from the plastic mariculture facilities, suggesting that mariculture makes a significant contribution to microplastic pollution in Sanggou Bay. Estimated inventory of microplastics in surface sediments of Sanggou Bay was 183.73 tons. Our results improve the understanding of risks caused by mariculture-derived microplastics to marine ecosystem and human health.

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1. Introduction

Plastic products are widely applied in various aspects of daily life because of their durability and affordability. In 2010, an estimated 4.8–-12.7 million tons of plastics arrived in the ocean from land-based sources in 192 coastal countries (Jambeck et al., 2015). Microplastics (defined as plastic particles ˂ 5 mm) account for the majority of plastic debris (Arthur et al., 2009; Van Cauwenberghe et al., 2013) and mainly originate from two sources: micron-sized plastic particles produced in the commercial manufacture (primary microplastics) and fragmentation of larger plastic debris already present in the marine environment (secondary microplastics) (Cole et al., 2011). Vast quantities of microplastics have accumulated in the ocean for long periods of time owing to their resistance to degradation (Cooper and Corcoran, 2010; Yoshida et al., 2016). It was estimated that 15–51 trillion microplastic fragments with a total weight of 93–236 thousand tons were present in the global ocean by the end of 2014 (Sebille et al., 2015). Because of their small size, microplastics can be readily ingested by marine organisms, causing physical and mechanical damage on them (Yin et al., 2018; Cong et al., 2019). In addition, microplastics may act as carriers of adsorbed pollutants and enhance their bioavailability, thereby inducing greater toxicity to marine organisms (Lu et al., 2018; Xia et al., 2020). Microplastics can also be transferred to marine organisms at higher trophic levels and even humans through the food chain (Carbery et al., 2018). Therefore, marine microplastic pollution has been declared as one of the most critical issues by the United Nations Environment Programme (UNEP, 2016).

Microplastics have different densities based on their polymer type (Imhof et al., 2012). On the one hand, the microplastics that are denser than seawater will sink into the sediments. On the other hand, microplastics that are less dense than seawater eventually settle because they interact with aggregates, biofouling agents, and faecal matter, which increase their density (Koelmans et al., 2017; Rummel et al., 2017; Porter et al., 2018). Furthermore, the upward transport of buried microplastics by bioturbation is negligible (Näkki et al., 2019). Approximately 99% of the plastic entering the ocean, including buoyant polymers, will ultimately settle in sediments (Koelmans et al., 2017). Therefore, marine sediments are expected to be the ultimate sink for microplastics (Ivar et al., 2014). To date, some studies have reported microplastic pollution in sediments of the Bohai Sea (Zhao et al., 2018), Northern Yellow Sea (Zhu et al., 2018), South Yellow Sea (Wang et al., 2019a) and Jiaozhou Bay (Zheng et al., 2019). However, there are limited reports quantifying the inventory of microplastics in marine sediments.

Mariculture has great potential to improve human nutrition through providing seafood globally (Liu et al., 2018). Approximately one-third of the total global seafood production was supplied by mariculture in 2016 (FAO, 2018). The development of mariculture has heavily relied on plastic facilities such as fishing nets, ropes, buoyant material, and net cages, which is unlikely to change in the near future. Under direct UV light, wave action, abrasion and physical breakdown for a long time, the plastic facilities will inevitably be a considerable source of microplastics in mariculture areas (Chen et al., 2018; Krüger et al., 2020). China is the world's largest mariculture producer, accounting for >17% of the global mariculture volume in 2014 (FAO, 2017). Sanggou Bay is one of the most important mariculture regions in China and nearly the whole bay is covered by mariculture facilities. Wang et al. (2019b) reported that microplastic levels in the seawater, sediment, and oysters from Sanggou Bay in 2017 were 63.6 $\pm$ 37.4 items/L, 2178 $\pm$ 369 items/kg, and 41.0 $\pm$ 15.5 items/individual. However, the spatiotemporal distribution of microplastics in the sediments of Sanggou Bay and the contribution of mariculture facilities to the microplastics levels remain unclear. In this study, we aimed to: (1) investigate the spatiotemporal distribution of microplastics in surface sediments of Sanggou Bay, including abundance, size, shape, color and polymer type; (2) identify the contribution of mariculture facilities to microplastics in surface sediments from Sanggou Bay; and (3) estimate the inventory of microplastics in surface sediments. To our knowledge, this is the first study to investigate the source and inventory of microplastics in sediments of Sanggou Bay.

2. Materials and methods

2.1. Study area

Sanggou Bay (37°01′–37°09′N, 122°24′–122°35′E), a typical semi-closed bay in the northern part of the Yellow Sea, is located on the eastern side of the Shandong Peninsula. Sanggou Bay covers 150.3 km^2, with an average water depth of 7.5 m (Jiang et al., 2015). The tide of Sanggou Bay is irregular semidiurnal. The Sanggou, Yatou, and Gu Rivers flow into Sanggou Bay, with an annual water discharge of $(1.7–2.3) \times 10^8$ m^3. Sanggou Bay has been used for mariculture for >30 years and has become one of the most important aquaculture regions in China (Xia et al., 2019). The main cultured species include kelp (*Laminaria japonica*), scallop (*Chlamys farreri*), oyster (*Crassostrea gigas*), fish (*Paralichthys olivaceus*) and so on. Bivalves are mostly raised in cages hung from rafts, kelp is tied to ropes and grows downward in the water column, and fish are cultured in submerged net cages. Sanggou Bay serves as a major source of seafood for the local people.

2.2. Sample collection

Surface sediment samples were obtained in triplicate from 8 sites in November (autumn) 2015, March (winter), June (spring) and August (summer), 2016 in Sanggou Bay (Fig. 1). In brief, stations 1, 4, and 7 are located in the coastal waters, stations 2, 5, and 8 are located in the centre of the bay, and stations 3 and 6 are located in the mouth of the bay. A Van Veen grab (HYDRO-BIOS, Germany) was used to obtain samples of the surface sediment (0–2 cm). Subsequently, the samples were transferred to an aluminium foil sampling bag with a stainless-steel spatula and immediately stored in an incubator filled with ice cubes until analysis. To avoid contamination by other plastic materials during samples collection and storage, only glass and aluminium instruments were employed. In addition, we collected the plastic aquaculture facilities in Sanggou Bay, including fishing buoys, fishing rope and foam, to identify their polymer types.

2.3. Extraction of microplastics

Extraction of the microplastics was performed according to Zhu et al. (2018), with some minor modifications. In the laboratory, all glassware was rinsed with distilled water to ensure that no plastic contamination occurred during the experimental period. Each sample was weighed to approximately 300 g, dried in an oven, and weighed again in a glass beaker. Then, 250 mL of saturated NaCl solution was added and stirred for 3 min to ensure that the microplastic was suspended. After 12 h of settlement, the supernatant liquid was filtered through a 30 μm steel sieve using a vacuum suction device and washed repeatedly with distilled water to remove NaCl. The material on the membrane was transferred to a glass beaker, 30% H_2O_2 was added to remove organic matter, and the material was suspended in a saturated NaCl solution. The supernatant liquid was filtered using a 30 μm steel sieve and washed with distilled water at least five times to remove NaCl. Microplastics were transferred from the steel sieve to Petri dishes for microscopic examination.

2.4. Observation and identification

Microplastics were counted and photographed by optical microscopy (Olympus BX-51, Japan). According to the identification rules of previous studies, microplastics were classified into six size grades: <0.5 mm, 0.5–1 mm, 1–2 mm, 2–3 mm, 3–4 mm, and 4–5 mm; shape

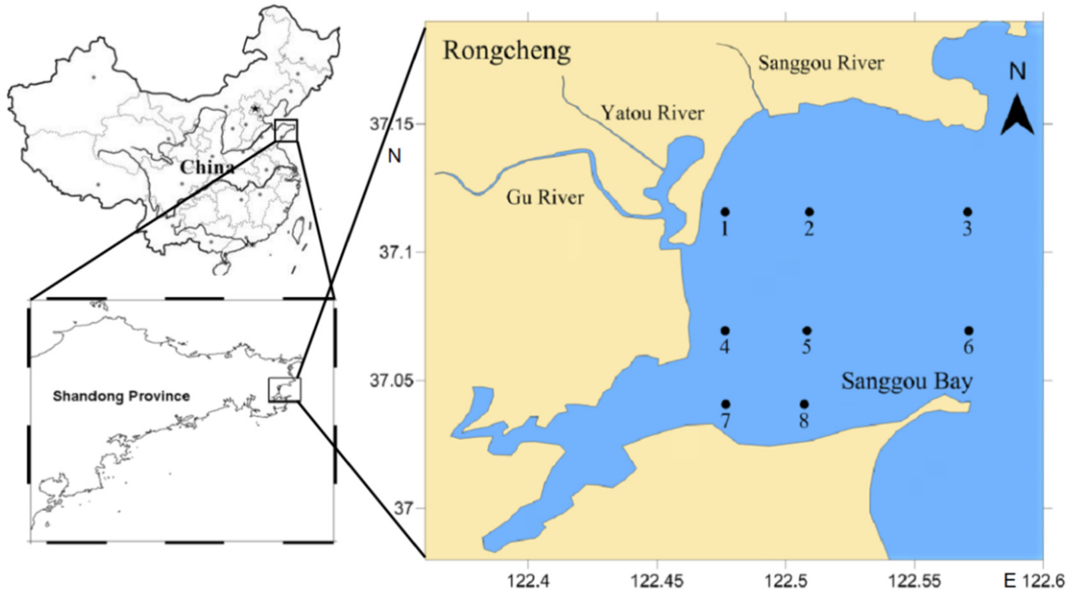

Fig. 1. Locations of sampling sites in Sanggou Bay, China.

was divided into film, granule, fibers and pellet; and color was divided into black, transparent and other colors. The composition of suspected plastics was identified by micro-Fourier Transform Infrared Microscope (micro-FTIR; Thermo Fisher Nicolet iN10, USA) (Jabeen et al., 2016). The micro-FTIR spectrum of each plastic item was recorded between 4000 and 650 cm^{-1} at a resolution of 8 cm^{-1} using a collection time of 3 s and with 16 co-scans. Each spectrum was compared with the OMNIC standard spectral library. When the matching degree is higher than 70%, suspected particles can be identified as microplastics (Yu et al., 2016). The actual number of microplastics was recalculated based on the micro-FTIR analysis. In addition, the plastic mariculture facilities, including buoys, fishing rope, and foam, were fragmentized to identify the polymer composition using the micro-FTIR spectrum to analyse the contribution of mariculture to microplastic pollution in sediments.

2.5. Sources of microplastics

It has been reported that the morphology and polymer composition of microplastics can be used as key indicators for source identification of microplastics (Lei et al., 2019). To identify the sources of microplastics, we compared the microplastics in sediments with the four types of mariculture facilities based on morphology combined with polymer

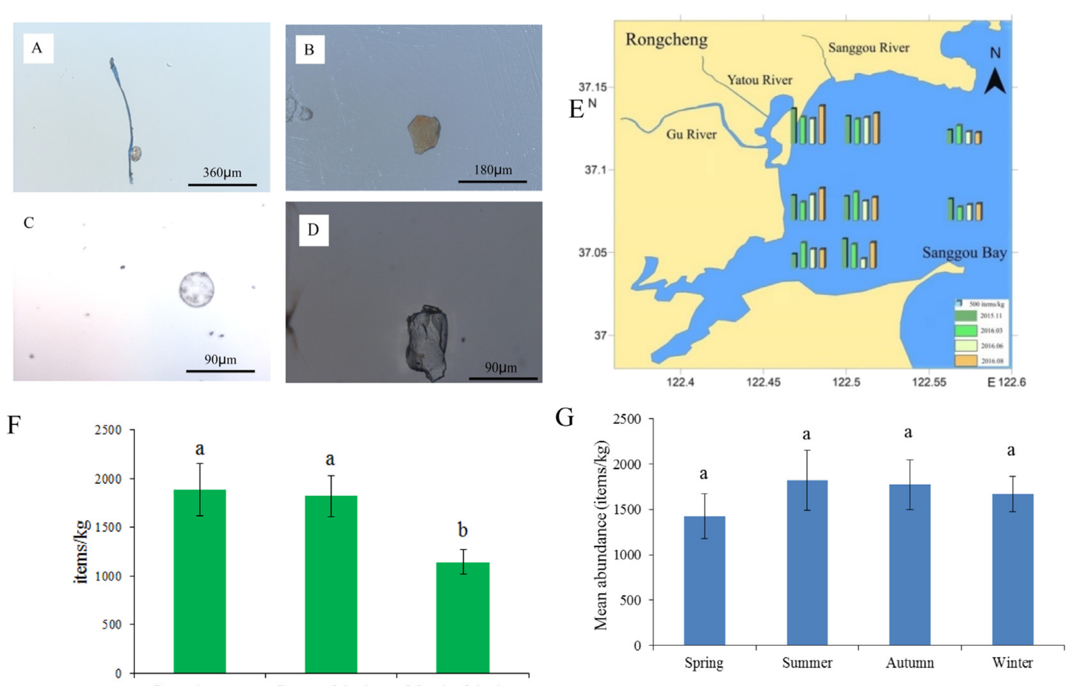

Fig. 2. Representative photographs of microplastics by stereo microscope. (A) Fibers, (B) Film, (C) Pellet, (D) granules. (E) Abundance and spatiotemporal distribution of microplastics. (F) Abundance of microplastics in coastal waters, centre and mouth of the Sanggou Bay. (G) Abundance of microplastics in spring, summer, autumn and winter. Different letters show significantly differences among different areas ($p < 0.05$).

Table 1
A summary of microplastic pollution in sediments of mariculture areas, coastal waters, estuaries, and open sea worldwide.

Location	Size	Abundance (items/kg dw)	Shape	Color	Composition	Reference
Mariculture areas						
Xiangshan Bay, China	333 μm–5 mm	1739 ± 2153	Fibers	/	PE, synthetic cellulose	Chen et al., 2018
Sanggou Bay, China	50 μm–5 mm	2178 ± 369	Fibers, granules, lines, films, fragments, spherules	Yellow, red, transparent, black, white, blue	/	Wang et al., 2019b
Sishili Bay, North Yellow Sea, China	35 μm–4.985 mm	499.76 ± 370.07	Fibers	Transparent, red	Rayon, PE, PP	Zhang et al., 2019
Sanggou Bay, China	30 μm–5 mm	1674.4 ± 525.6	Fibers, film, pellet, granules	Transparent, Black, other color	PS, PE, PC, PP, cellulose	This study
Coastal waters						
Victoria Harbour, Hong Kong	0.1 mm–4.7 mm	263 ± 83	Fragment, pellet, line fibers	Blue, brown, white, green, transparent, orange, yellow, red, pink, grey, dark	PP, PE, SAN	Tsang et al., 2017
Jiaozhou Bay, China	0.45 μm–4 mm	15 ± 6	Fibers, fragments, granules	Black, blue, red, green	PET, PE, PP, PA, CP, PVC, PS, LDPE	Zheng et al., 2019
Bay of Brest, Brittany, France	1.6 μm–5 mm	0.97 ± 2.08	Fragments, granules	Blue, transparent	PE, PP, PS	Frère et al., 2017
Harbour, Belgian Coast	38 μm–1 mm	166.7 ± 92.1	Fibers, granule		Nylon, PVA, PP	Claessens et al., 2011
Estuaries						
Changjiang Estuary, China	1 μm–5 mm	121 ± 9	Fibers, fragment, granule	Transparent, blue, black, yellow, red, white	Rayon, polyester, acrylic	Peng et al., 2017
Jagir Estuary, Surabaya City, Indonesia	0.3 mm–5 mm	345.2 ± 216.6	Fibers, fragment, film	Transparent, blue, black, yellow, red, white	PP, PES, LDPE	Muhammad et al., 2019
Open sea						
Bohai Sea, China	1 μm–5 mm	171.8 ± 55.4	Fibers, granule	Transparent	PP, PE, PS	Zhao et al., 2018
Southern Yellow Sea, China	1 μm–5 mm	72.0 ± 27.2	Fibers	/	Rayon	Zhao et al., 2018
North Yellow Sea, China	30 μm–5 mm	37.1 ± 42.7	Film, fiber, granule	Transparent, black, other color, white	PE, PP, PE/EA, PP/PE, nylon	Zhu et al., 2018

Note: PE: polyethylene; LDPE: low-density polyethylene; PP: polypropylene; PS: polystyrene; PC: polycarbonate; PA: polyamide; PVC: polyvinyl chloride; PES: polyether sulfone; PVA: polyvinyl alcohol; CP: cellophane; PET: polyethylene terephthalate; SAN: styrene acrylonitrile.

composition analysis by Chen et al. (2018). In brief, morphological comparisons were performed based on the color and shape analysis between microplastics and mariculture facilities. Polymer compositions of microplastics were compared with those of mariculture facilities. Table S1 shows the criteria for source determination of microplastics. Microplastics conforming to both morphological and polymer composition were identified as mariculture-derived microplastics.

2.6. Inventory of microplastics

According to the method by Cai et al. (2018), with some minor modification, the inventory of microplastics in surface sediments of Sanggou Bay was calculated as follows:

$$I_{ix} = \sum_{i=1}^{3} C_i S_i H_x \rho_i$$

$$I = (I_{ix} \times V_a \times \rho_a)/1000.$$

where I_{ix} represents the inventory (items), C_i represents the microplastic abundance (items/kg) of sediment in different areas, S_i represents the different sea area (m^2), it accounts for one quarter, one half and one quarter of the breeding area in Sanggou Bay, H_x is the depth of surface sediments (2 cm), ρ_i represents the density (kg/m^3) of sediment in different areas was calculated using ring tool method. The term i represents the different areas, including the coastal waters, the centre of the bay and the mouth of the bay, respectively. I represents the inventory (tons). Finally, V_a represents the average volume (m^3) of microplastics, and ρ_a is the average density (kg/m^3) of microplastics.

2.7. Quality assessment and quality control

Throughout the entire sample collection process, sample separation and sample analysis, we performed precautions to avoid contamination by external plastics. In brief, all glass containers were soaked with dilute nitric acid overnight and then rinsed three times with deionized water. All chemical reagents were filtered through 0.45 μm glass membranes prior to use. Microscopic examination was performed in the laboratory with closed windows to minimize air pollution. Cotton clothing, gloves and face masks were worn during field sampling and analytical procedures to prevent contamination by external synthetic fibers. Solution preparation and extraction of microplastics were always performed in a laminar flow cabinet. Procedural blanks (only distill water without field water or sediment) were run within the same procedure to ensure no plastic contamination during the analysis.

2.8. Data analysis

The microplastic abundance in sediment samples was expressed in terms of items per unit mass of dry sediment (items/kg dw). Each sediment sample was performed in triplicate, and the microplastic abundance was expressed as mean values ± standard deviation (SD). One-way ANOVA, followed by Tukey's range tests, was performed to compare the microplastics abundance among the different regions and seasons using the IBM SPSS statistics programme (version 22.0). Prior to analysis, Levene's and Shapiro–Wilk's tests were used to verify the homogeneity and normality of variance, respectively. Differences were considered significant at $p < 0.05$.

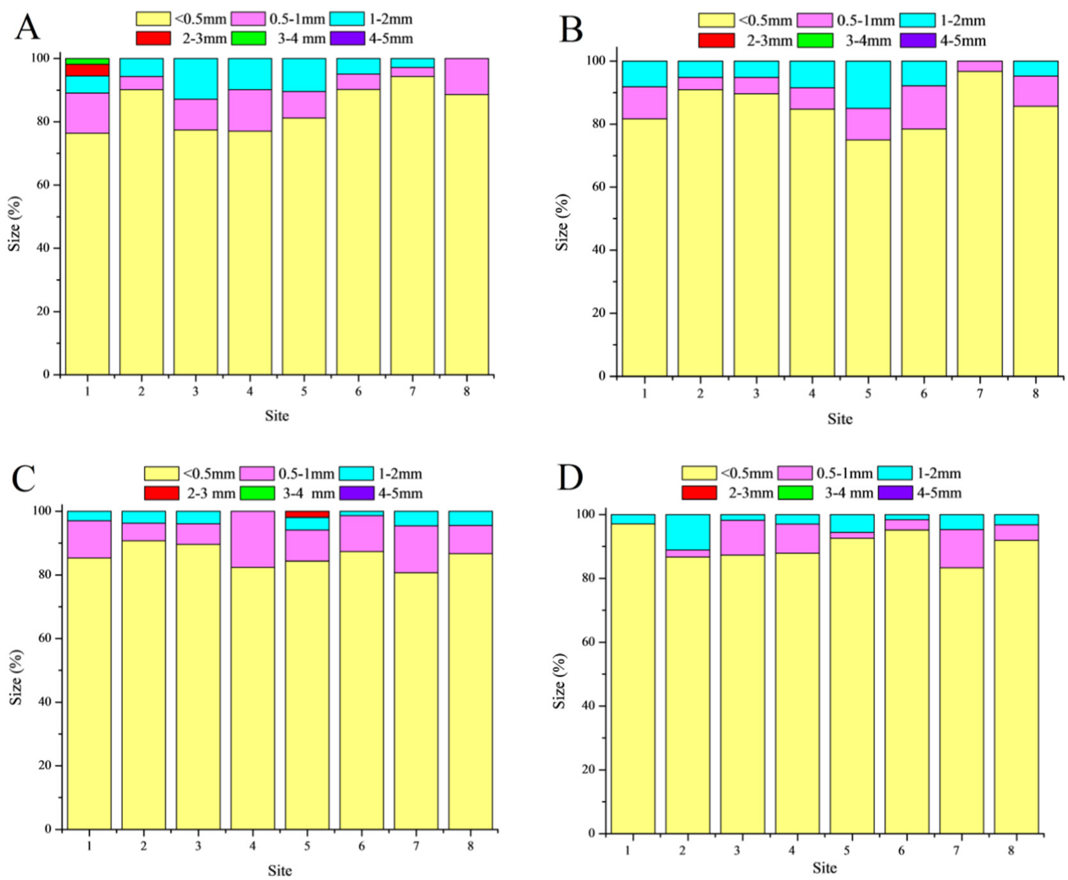

Fig. 3. Size distributions of microplastics collected from surface sediments of Sanggou Bay in (A) spring, (B) summer, (C) autumn, and (D) winter.

3. Results and discussion

3.1. Abundance and spatiotemporal distribution of microplastics

No microplastics were found on the blank controls. Therefore, no blank correction was needed. Microplastics were detected in all the surface sediment samples, with a total number of 8748 pieces in all four seasons. Representative photographs of microplastics are shown in Fig. 2A, B, C and D. The abundance of microplastics in surface sediments ranged from 699 items/kg to 2824 items/kg, with higher abundance within the bay and lower abundance at the mouth of the bay (Fig. 2E). The average abundance of microplastics in the surface sediments was 1674 ± 526 items/kg dw, which was similar with the previous results in Sanggou Bay (Wang et al., 2019b). Microplastic abundance in the sediments from mouth of the bay (1142 ± 254 items/kg) were significantly lower than that in the centre of the bay (1818 ± 419 items/kg) and coastal waters (1885 ± 534 items/kg) (Fig. 2F). The higher abundance of microplastics within the bay may be caused by terrestrial runoff and mariculture activities, and the lower abundance of microplastics in mouth of the bay may be the result of the strong current speed, leading to the poor settlement of the microplastics. For the seasonal distribution, the maximum abundance of microplastics in the surface sediments appeared in summer (1824 ± 659 items/kg), followed by autumn (1775 ± 546 items/kg), winter (1673 ± 387 items/kg) and spring (1426 ± 483 items/kg). However, there were no significant differences among the four seasons (Fig. 2G). Therefore, significant differences for microplastics abundance were observed in the spatial distributions rather than the temporal distributions.

As shown in Table 1, some studies have investigated the microplastic pollution in marine sediments. The isolation methods of microplastics in sediments are relatively uniform, including density separation, digestion, and filtration. Therefore, we compared our results with those from the previous studies. The microplastic pollution level in the sediments of Sanggou Bay was slightly lower than that in Xiangshan Bay, which is a typical farming bay. However, they had the same order of magnitude (Chen et al., 2018). The abundances of microplastics in the sediments of Sanggou Bay were higher than those of the estuaries (e.g., Changjiang Estuary and Jagir Estuary) (Peng et al., 2017; Muhammad et al., 2019), coastal waters (e.g., Victoria Harbour, Jiaozhou Bay, Brest Bay, and the Belgian Coast) (Claessens et al., 2011; Frère et al., 2017; Tsang et al., 2017; Zheng et al., 2019), and open sea (e.g., Bohai Sea, North Yellow Sea and South Yellow Sea) (Zhao et al., 2018; Zhu et al., 2018). The higher microplastics pollution in mariculture areas (e.g., Sanggou Bay and Xiangshan Bay) may be caused by the large number of plastic culture facilities and weaker hydrodynamic forces. Above all, the microplastic pollution in the surface sediments of Sanggou Bay represented a heavy level. On the one hand, because of their large surface area and hydrophobic nature, microplastics can adsorb various environmental pollutants (Wang et al., 2018). Furthermore, the adsorption capacities of microplastics were enhanced with the increase of aging degree (Ding et al., 2020). Therefore, a large number of microplastics buried in surface sediments ought to cause serious threat to marine benthic organisms. On the other hand, microplastics incorporated into marine snow are deposited into the marine sediments (Porter et al., 2018), which will influence the biogeochemical cycle in coastal waters.

3.2. Characteristics of microplastics in sediments of Sanggou Bay

As shown in Figs. 3 and S1, microplastics with a size of <0.5 mm were dominant in the sediments of Sanggou Bay, accounting for 76.36–94.37% in spring, 75.00–96.67% in summer, 80.68–90.74% in

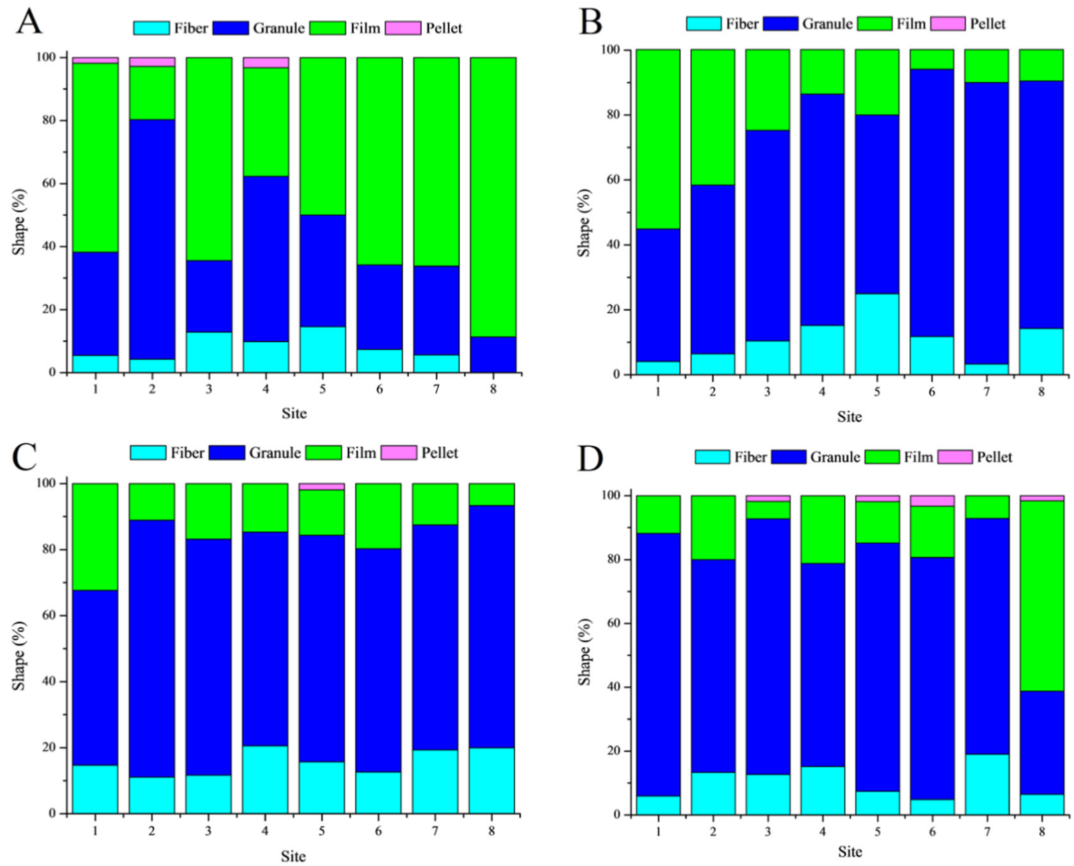

Fig. 4. Shape distributions of microplastics collected from surface sediments of Sanggou Bay in (A) spring, (B) summer, (C) autumn, and (D) winter.

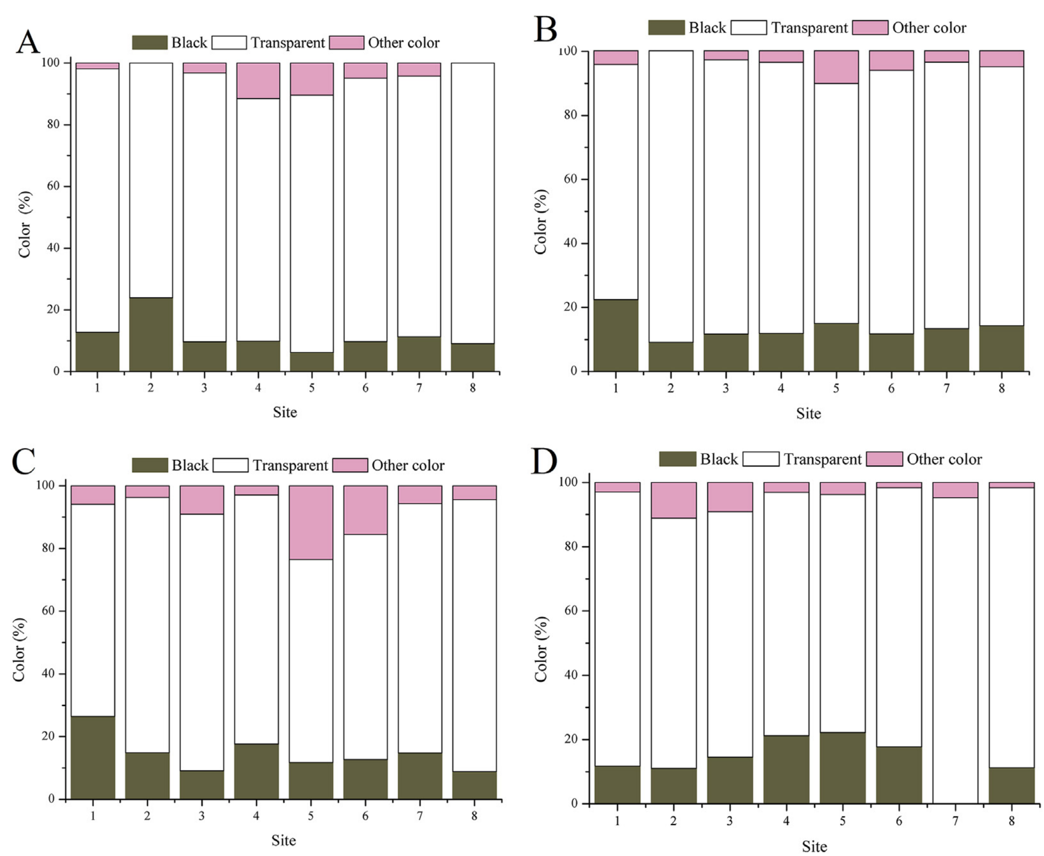

Fig. 5. Color distributions of microplastics collected from surface sediments of Sanggou Bay in (A) spring, (B) summer, (C) autumn, and (D) winter.

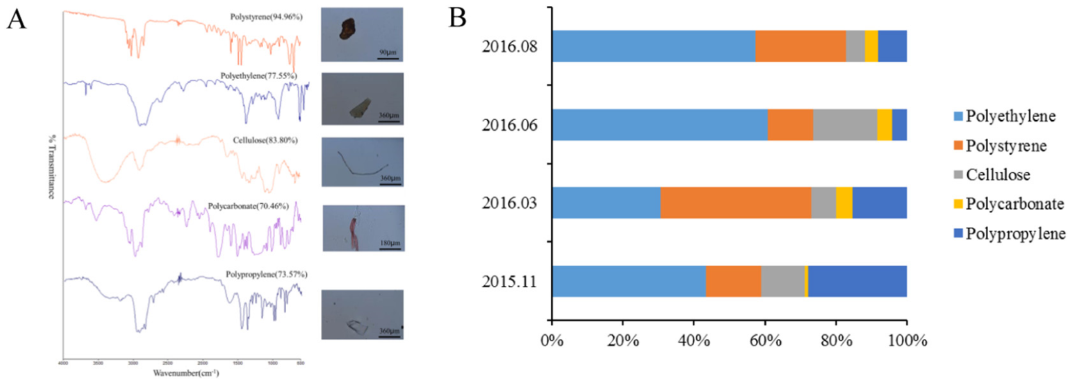

Fig. 6. Compositions of microplastics collected from surface sediments from Sanggou Bay: (A) photographs of typical microplastics and micro-FTIR spectra with their match degrees with the standard spectra and (B) proportion of polymer types over four seasons.

autumn and 83.33–97.06% in winter of the total number of microplastics, whereas microplastics with a size of >2 mm were scarce. In terms of spatial distribution, microplastics with a size of <0.5 mm were dominant in the sediments of Sanggou Bay in three areas, accounting for 76.36–97.06% in coastal waters, 75.00–92.59% in the centre of the bay and 77.42–95.16% in the mouth of the bay. The results are consistent with previous studies (Wang et al., 2019a). Under mechanical action, photooxidation, and biodegradation, the large plastic materials are ultimately fragmented into microplastics in marine environments (Gewert et al., 2015). Consequently, the number of small microplastics can increase exponentially (Isobe et al., 2015). Importantly, it has been reported that the smaller microplastics can induce higher toxicity to monogonont rotifer *Brachionus koreanus* (Jeong et al., 2016). In addition, the microplastic has larger specific surface area with the decrease of particle size, absorbing more pollutants and causing greater toxicity to marine organisms (Xia et al., 2020). Therefore, the occurrence of small microplastics in mariculture areas can pose more threat to marine environments.

As shown in Figs. 4 and S2, the dominant shape of the microplastics was granule over three seasons, accounting for 40.82–86.67% in summer, 52.94–77.78% in autumn, and 32.26–82.35% in winter. The majority shape of microplastics in spring was film, accounting for 16.90–88.64% of the total microplastics. In terms of spatial distribution, except the spring, the granule-shaped microplastics were dominant in the sediments of Sanggou Bay in the different areas, accounting for 40.82–86.67% in coastal waters, 32.26–77.78% in the centre of the bay, and 64.94–82.35% in the mouth of the bay. In general, spherical microplastics are primary microplastics, while film, fiber, and granule microplastics are secondary microplastics. Therefore, the secondary microplastics were dominant in sediments of Sanggou Bay. Similarly, the dominant shape of microplastics in Baltic Coast German was granule, followed by fibers (Stolte et al., 2015).

As shown in Figs. 5 and S3, the color of microplastics in surface sediments was mainly transparent, accounting for 76.06–90.91% in spring, 73.47–90.91% in summer, 64.71–86.67% in autumn, and 74.07–95.24% in winter. In terms of spatial distribution, microplastics with a color of transparent were dominant in sediments of Sanggou Bay, accounting for 67.65–95.24% in coastal waters, 64.71–90.91% in centre of the bay and 71.83–87.10% in mouth of the bay, respectively. The proportion of black plastics was 0–26.47% at all stations in four seasons. Colored plastics included red, yellow, green, purple, brown, and green, and accounted for 0–23.53% at all stations in the four seasons. Some marine predators may mistakenly ingest microplastic that is similar to the color of their prey (Peters et al., 2017). For instance, ingested pieces in larval fish from surface slicks were primarily blue or translucent in color, possibly because larval fish confuse the thread-like ocean colored plastic particles for copepod antennae (Gove et al., 2019).

In this study, 5000 suspected microplastics (57.2% of all surface sediment particles) were randomly selected and analysed by micro-FTIR. As shown in Fig. 6A, five component types of microplastics were identified in the sediments of Sanggou Bay, including polyethylene (PE), polystyrene (PS), polypropylene (PP), polycarbonate (PC) and Cellulose. The PE was the dominant compositions in three seasons, accounting for 60.64% in summer, 57.27% in autumn, and 43.33% in winter, respectively. In spring, PS accounted for the largest proportion of 42.35%, followed by PE (30.59%) and PP (15.29%) (Fig. 6B). In general, PE, PS, and PP were the main polymer type in sediments of Sanggou Bay, supported by Wang et al. (2019b), who found that PE, PP and PS were the main compositions of microplastics in the surface seawater of Sanggou Bay. Although the PE, PP and PS were less dense than the seawater, the biofouling and biological ingestion-excretion adsorbed to microplastics can enhance the density of plastics debris, causing them to settle onto the surface sediments (Katija et al., 2017; Kooi et al., 2017; Porter et al., 2018).

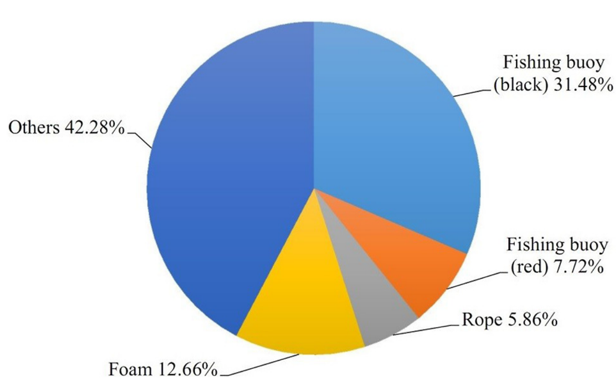

Fig. 7. Contribution rates of fishing buoy (black), fishing buoy (red), rope and foam to microplastics in surface sediments of Sanggou Bay.

3.3. Source identification of microplastics in sediments of Sanggou Bay

Based on micro-FTIR analysis, the polymers types of plastic mariculture facilities (e.g. fishing buoy, fishing rope, and foam) were depicted in Table S2. The fishing buoys and rope were mainly composed of PE, and the PS was the dominant composition of foam, which was consistent with the polymers types of the microplastics in the surface sediments of Sanggou Bay. According to the criteria for source determination of microplastics (Table S1), the contribution rates of fishing buoys (black), fishing buoys (red), rope, and foam to the microplastics in the surface sediments of Sanggou Bay were 31.48%, 7.72%, 5.86% and 12.66%, respectively (Fig. 7). Therefore, approximately 57.72% of microplastics in surface

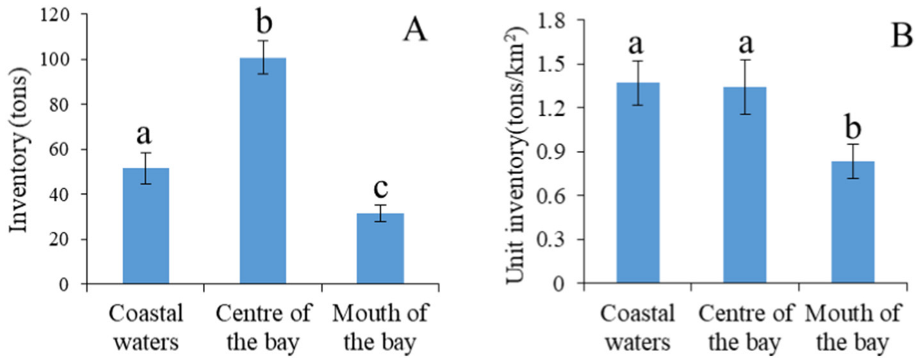

Fig. 8. Inventory of microplastics in surface sediments of Sanggou Bay. (A) Total inventory and (B) inventory per square kilometre. Different letters denote significant differences among different areas ($p < 0.05$).

sediments of Sanggou Bay originated from the plastic mariculture facilities, which was closely related to the raft culture model of Sanggou Bay. Sanggou Bay is a typical mariculture region in China with intensive suspended rafts, where an estimated 7 million plastic floating buoys exist (Fig. S4A). Under the influence of mechanical effect, biodegradation and photodegradation, the plastic facilities in Sanggou Bay breaks down into a large number of microplastics. In situ observation revealed that some fouling organisms (e.g., oysters and barnacles) were attached to the floating buoys (Fig. S4B). It has been reported that boring sphaeromatidae isopods destroyed expanded polystyrene foam floats under docks, and accelerated the fragmentation of plastics (Davidson, 2012). A single sea urchin *Paracentrotus lividus* produced on average 91.7 plastic fragments over 10 days (Porter et al., 2019). Similarly, approximately 55.7% and 36.8% of the microplastics in the surface seawater and sediment of Xiangshan Bay derived from different plastic gear, respectively (Chen et al., 2018). The dominant microplastics in surface sediment of Beibu Gulf originated from the abrasion of fishing gear, which contributed to 61.6% of the total abundance of microplastics (Xue et al., 2019). Therefore, the mariculture facilities make a considerable contribution to the microplastics in the mariculture areas.

3.4. Inventory of microplastics in sediments of Sanggou Bay

According to the percentage of microplastics with different sizes, we calculated the average size of microplastics as 0.25 mm. In addition, we defined the average density of microplastics as 0.98 g/cm^3 according to the density of PE and PS, which were dominant polymer of microplastics in sediments. The inventory of microplastics in the surface sediments of Sanggou Bay was estimated to be 1.20×10^{13} particles, equivalent to weight of 183.73 tons. Similarly, Cai et al. (2018) estimated that the inventory of microplastics in the surface seawater of the entire South China Sea was 700 tons. Xue et al. (2019) estimated that the storage of microplastics in deep sediments (−5 to −60 cm) and surface sediments of Beibu Gulf was 185 tons and 37 tons, respectively. Therefore, the inventory of microplastics in surface sediments of Sanggou Bay was comparable to that in deep sediments of Beibu Gulf. Bioturbation might be responsible for the vertical transport of "fresh microplastics" to "old sediment". For instance, Lugworm (*Arenicola marina*) can promote unidirectional transport of microplastics from surface sediments to deep sediments (−20 cm) (Gebhardt and Forster, 2018). Therefore, a huge amount of microplastics are preserved in sediments of Sanggou Bay. In terms of spatial distribution, the largest inventory of microplastics was located in the centre of the bay (100.85 tons), followed by the coastal waters (51.54 tons), and the least was in the mouth of the bay (31.34 tons). Furthermore, there were significant differences among the three areas (Fig. 8A). Therefore, the centre of the bay was the largest reservoir of microplastics in Sanggou Bay. In addition, the unit inventories of microplastics in the coastal waters (1.37 tons/km^2) and the centre (1.34 tons/km^2) were significantly higher than that in the mouth of the bay (0.84 tons/km^2) (Fig. 8B), which can be explained by the strong current speed in the mouth of Sanggou Bay. In future studies, it will be crucial to obtain vertical profile of microplastic inventory in ocean to yield the realistic microplastic load in various compartments (e.g. seawater and sediments) of marine environments.

4. Conclusions

In this study, we investigated the spatiotemporal pollution patterns of microplastics in the surface sediments of Sanggou Bay. The microplastics were mainly smaller debris, and transparent in color. Granule and film were the dominant shape in all samples. The dominant polymer was PE in summer, autumn, and winter, and PS in spring. When compared the results with other sea areas (e.g., coastal waters, estuaries, open sea and other mariculture areas), the microplastic pollution in surface sediments of Sanggou Bay was at a heavy level. The source identification of microplastics analysis suggested that the plastic debris mainly originated from the mariculture facilities due to intensive raft culture. The inventory of microplastics in the surface sediments from Sanggou Bay was estimated to be 183.73 tons. It is worth noting that we only estimated their masses of microplastics with size larger than 30 μm, indicating that microplastics smaller than 30 μm were generally missed. Therefore, the actual inventory of microplastics in the surface sediments of Sanggou Bay might be underestimated. The high levels of microplastic pollution in mariculture areas can cause a potential risk to seafood safety and even human health. Further research is strongly needed to assess the quality and safety of seafood in mariculture areas.

CRediT authorship contribution statement

Qi Sui: Conceptualization, Investigation, Writing - original draft. **Longjun Zhang:** Data curation, Resources. **Bin Xia:** Investigation, Writing - review & editing, Supervision, Resources, Funding acquisition. **Bijuan Chen:** Data curation, Validation. **Xuemei Sun:** Data curation, Software. **Lin Zhu:** Methodology. **Rongyuan Wang:** Data curation. **Keming Qu:** Resources, Supervision.

Declaration of competing interest

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

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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.2020.138064.

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