Marine Pollution Bulletin 176 (2022) 113436

Contents lists available at ScienceDirect

Marine Pollution Bulletin

journal homepage: www.elsevier.com/locate/marpolbul

Characteristics and spatiotemporal distribution of microplastics in sediments from a typical mariculture pond area in Qingduizi Bay, North Yellow Sea, China

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ARTICLE INFO

Keywords:
Microplastics
Mariculture pond
Sediments
Spatiotemporal distribution
Risk assessment

ABSTRACT

Microplastics (MPs) in mariculture environments may have an impact on mariculture and ecosystems. This study sampled the sediments in mariculture ponds and offshore areas in Qingduizi Bay during winter and summer. The abundance, characteristics, spatiotemporal distribution and pollution risk of microplastics were analyzed. The results showed that the abundance of MPs in the mariculture pond and offshore area was 49.2 ± 35.9 items·kg^{-1} d.w. and 17.1 ± 9.9 items·kg^{-1} d.w.; the MPs were mainly composed of transparent fibers of thickness 2000–5000 μm, with the main polymers being polyethylene terephthalate (PET) and cellophane (CP). The spatial distribution showed a downward trend from the inside to the outside, but the difference was not significant when comparing different seasons. The pollution load index (PLI) risk assessment showed that all sampling sites were at Hazard Level I. This study can provide valuable information for the risk assessment of microplastic pollution in mariculture areas.

1. Introduction

Microplastics (MPs) (plastic <5 mm in diameter) are considered a "new emerging pollutant" and have attracted extensive attention in various fields (Blair et al., 2017; Jambeck et al., 2015; Rochman, 2018; Thompson et al., 2004). At present, it is generally believed that MP pollution is a threat to the global ecological environment (Rochman and Hoellein, 2020). Therefore, a series of studies were carried out to investigate MP pollution in marine environments, such as seafloor sediments (Abel et al., 2021), various seawater depths (Tekman et al., 2020), and different coastal locations (Chen et al., 2021a; Lin et al., 2021). The results showed that MPs had spread all over the world. MPs contain harmful additives (benzophenone-3, phenol-formaldehyde) (Na et al., 2021; Trestrail et al., 2020) and carry persistent organic pollutants and potential pathogens, such as benzo[*a*]pyrene, Vibrio, and Pseudomonas (Tang et al., 2020; Zhang et al., 2021b), functioning as compound pollutants. At the same time, MPs are similar in size to some marine organisms, so they can easily enter the food chain and transmit through ingestion and excretion (Wang et al., 2019c), thus causing harm to marine organisms and ecosystems.

Mariculture generally involves a variety of human activities, which bring large amounts of MP pollution. Plastics are often used to make fishing gear, such as fishing nets, fishing lines, cages and ropes (Zhang et al., 2021a). During benthic dredging and trawling, mechanical wear of fishing gear usually increases the content of fiber MPs in marine fishery waters (Chen et al., 2021b). Meanwhile, plastic bags and bottles,

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https://doi.org/10.1016/j.marpolbul.2022.113436
Received 28 January 2022; Accepted 4 February 2022
Available online 11 February 2022
0025-326X/

fishing gear and other plastic products abandoned in mariculture ponds are easily broken and degraded into smaller particles due to aging and weathering (Ma et al., 2020; Shim et al., 2018). In the process of production, transportation, storage and feeding, MPs can be introduced into feed and fish drugs and then released into mariculture ponds (Zhou et al., 2020). On the other hand, due to wind, tides and human disturbance, the MPs in mariculture ponds are likely to migrate to offshore areas. However, the spatiotemporal distribution of MPs in the sediments of mariculture ponds and offshore areas is not clear. Current studies have observed a variety of potentially harmful effects of MPs on marine biota (Rezania et al., 2018), and have showed that MPs threaten hatcheries in mariculture areas (Campos et al., 2021). At present, different degrees of MP pollution have been found in mariculture ponds. Mohsen et al. (2019) found that the abundance of MPs was 20–1040 items/kg in the sediments of eight sea cucumber farms along the Bohai Sea and the Yellow Sea in China. Qingduizi Bay, an important semi-closed estuary in the northern Yellow Sea (Compilation Committee for Survey of China Bays, 1991), is considered to be the most developed mariculture area in China. At present, research on Qingduizi Bay has mainly focused on organic pollutants, such as polychlorinated biphenyls, organochlorine pesticides (Yang et al., 2019), and phenolic endocrine-disrupting compounds such as nonylphenol and octylphenol (Wang et al., 2019c). However, research on MP pollution has not yet been carried out.

In this study, the sediments of mariculture ponds and offshore areas in Qingduizi Bay were sampled during summer and winter. Furthermore, we analyzed the spatiotemporal distribution of MPs in sediments. We also discussed the source and migration of MPs in sediments and carried out a risk assessment of MP pollution in this area. Our results can provide basic data for the assessment of MP pollution in mariculture areas in China and provide important information to support mariculture management strategies and promote the sustainable and healthy development of mariculture.

2. Materials and methods

2.1. Study area

Qingduizi Bay (39°41′59″ – 39°49′31″N, 123°11′41″ – 123°26′06″E) is an important estuarine bay in the northern Yellow Sea of China, with the Nanjing Peninsula to the east and the Heidao Peninsula to the west, which covers a tidal flat area of 130 km^2 (Editorial Board of China Bay Survey, 1991). This bay is a typical semienclosed bay with a sea area of up to 5384 km^2, a large area of which has been farmed since the 1980s (Liu et al., 2018). Mariculture ponds in seawater are used by the mariculture industry to cultivate marine aquatic economic animals and plants in coastal shallow water or tidal flats. The ponds are usually classified according to the species of culture, such as fish ponds and shrimp ponds. In Qingduizi Bay, the main cultured species include sea cucumber, jellyfish and razor clam (Wang et al., 2019b), and the area of the mudflat culture is 600 ha (Wu et al., 2011).

2.2. Sediment sampling

A total of 20 sampling sites were set up in the mariculture pond (P) and the offshore area (S) of the outer bay (no mariculture activities) of Qingduizi Bay (10 in P, 10 in S). The sampling sites are shown in Fig. 1. At each site, in summer (July) and winter (December) of 2012, a stainless steel grab sampler was used to collect undisturbed sediments at 0–5 cm of the seabed surface. At each sampling site, three replicate sediments were collected, and a total of 120 sediment samples were collected. The collected samples were fully mixed and carefully covered with aluminum foil. These samples were initially stored in a freezer until they were transported to the laboratory and kept at −20 °C for further analysis.

2.3. Separation of MPs from sediments

MPs were separated from sediments by the method described by Li et al. (2021a), and some improvements were made, using a saturated sodium bromide solution (NaBr, ρ = 1.37 g/cm^3) as the density separation solution. First, sodium bromide was dissolved in pure water to form saturated sodium bromide, and then it was filtered by a vacuum pump through 8 μm glass microfiber filter membranes (Q/IEFJ01-1997) to eliminate impurities and external MP contamination. The filtered saturated sodium bromide solution was stored in a conical bottle and covered with aluminum foil. After thawing at room temperature, the sediments were transferred to a clean beaker and dried in an oven at 60 °C to a constant weight. The 50 g sample was put into the 500 mL beaker. Then, the filtered saturated sodium bromide solution of 200 mL was added to the beaker, and the resulting mixture was stirred with a glass rod for 3 min to fully mix the sample with the sediment. The

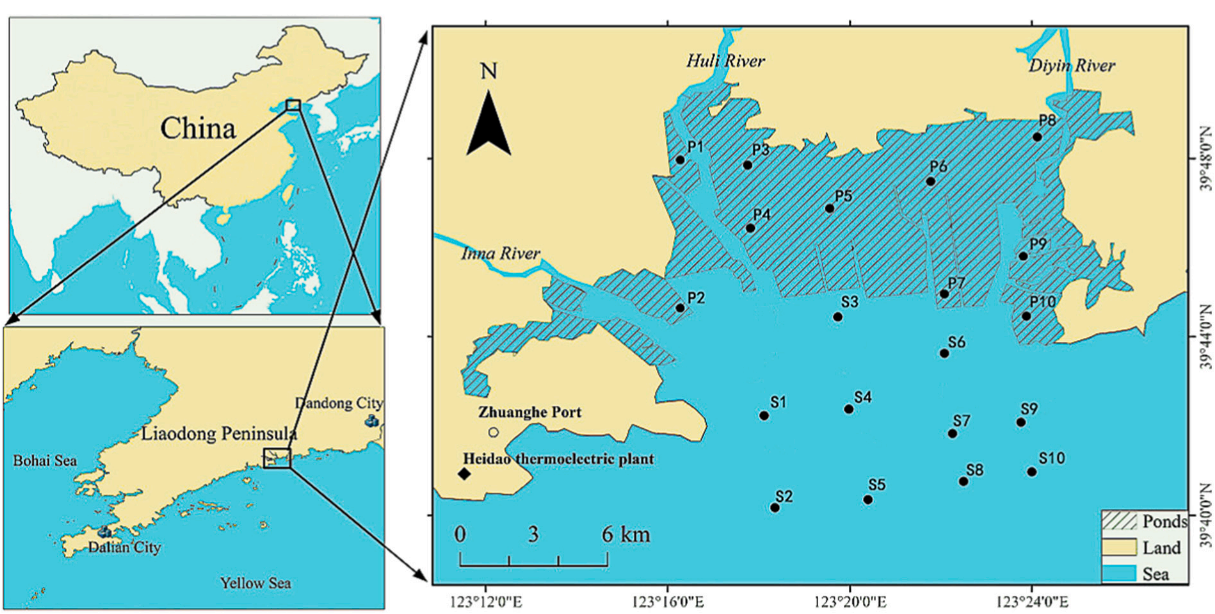

Fig. 1. The map of study area and sampling sites.

mixture was then left at room temperature for 2 h until the sediments were completely precipitated. Subsequently, the supernatant was transferred to a clean beaker, and the extraction step was repeated three times. Then, 5 mL H_2O_2 (30%) was added to the supernatant to digest organic substances in order to reduce the interference of impurities on subsequent tests. After standing for 24 h, the supernatant was filtered through an 8 μm glass fiber membrane (Q/IEFJ01-1997). At the same time, 200 mL of ultrapure water was used to wash the inner wall of the beaker and the pumping and filtration device to reduce the loss of MP samples caused by the adhesion of the experimental apparatus. The washed water was then filtered again. Then, the filter membrane was transferred to Petri dishes and air-dried naturally at room temperature.

2.4. MPs analysis

2.4.1. Visual identification

After air drying, all MPs on the filter membrane were carefully identified, photographed and counted with a Leica APO S-8 stereoscopic microscope equipped with a camera and a computer connection. The identification of MPs is based on their appearance and the following three selection criteria: (1) particles that cannot be torn or broken by tweezers, (2) particles without cell or organic structure, and (3) fibers that are equal in thickness throughout their entire length or that look like curved/twisted flat ribbons (Lin et al., 2021; Xue et al., 2020; Zhu et al., 2019). The shape, color and particle size of suspected MP particles extracted from sediment samples were characterized.

2.4.2. Polymer identification

MPs cannot be completely and accurately identified by visual observation. After the suspicious MPs were classified, the chemical composition of the suspected MP particles was identified and analyzed by Fourier transform infrared spectrometry (μ-FTIR).

The suspicious MPs were selected under a stereomicroscope, removed with tweezers, placed in a diamond compression cell (SPECAC V69108), and scanned and identified by Fourier transform infrared spectrometry.

Fourier transform infrared spectroscopy (Nicolet™ iN10, Thermo Fisher Scientific, USA) was performed. The parameters were set as follows: spectral range: 650–4000 cm^{-1}, spectral resolution: 8 cm^{-1}, and 128 scans for each measurement; the aperture size was adjusted according to the size of MPs, and the background was collected before sampling each time to reduce interference. The lower limit of the detected particle size for FTIR was 20 μm. OMNIC 8.2 software was used to analyze the absorption frequency of specific chemical bonds in related polymer samples; the spectrogram of polymer samples was obtained and was matched with the OMNIC polymer spectrum library to confirm the polymer type of MPs. Only when the matching degree between the spectrum and standard databases exceeded 70% could they be recognized as MPs (European Commission, 2013).

2.4.3. Quality assurance and quality control

To ensure the reliability of the results and reduce external MP pollution, strict quality control measures were taken while processing samples (Dehaut et al., 2019), including environmental pollution control, environmental background correction (Lin et al., 2021) and procedural background correction (Falahudin et al., 2020). The following pollution controls were adopted in this experiment: (1) In the process of sample preparation and analysis, only nonplastic samplers and containers were used, and they were washed with Milli-Q water three times before use and then covered with aluminum foil; (2) the investigators who processed the samples wore 100% cotton lab coats and latex gloves; (3) the windows were kept closed in the laboratory, and the whole experiment was carried out in a clean fume hood; (4) saturated sodium bromide solution (ρ = 1.37 g/cm^3) was filtered through an 8 μm glass fiber membrane (Q/IEFJ01-1997); (5) the environmental blank was set up, and a clean blank film was always placed near the sample during each batch of sample analysis to correct the potential environmental background; and (6) the procedural blank was set up using the same volume of saturated sodium bromide solution (ρ = 1.37 g/cm^3) with no sediment, and the same MP separation operation was carried out to correct the procedural background.

2.5. MPs risk assessment

The pollution load index (PLI) proposed by Tomlinson was used to evaluate the load of pollutants in the whole region (Tomlinson et al., 1980). The abundance of MPs was used instead of pollutant load to evaluate the pollution risk caused by MP in Qingduizi Bay (Kabir et al., 2021; Li et al., 2020a; Pan et al., 2021; Xu et al., 2018; Yan et al., 2021).

$$CF = C/C_0 \tag{1}$$

$$PLI_n = \sqrt{CF} \tag{2}$$

$$PLI_{zone} = \sqrt[n]{PLI_1 \times PLI_2 \times \cdots PLI_n} \tag{3}$$

In the formula, CF is the pollution coefficient of MPs, C is the measured content of MPs at a certain site, and C_0 is the reference value of MP concentration. C_0 is a defined value, and in this study, it is expressed as the lowest concentration of MPs in all sampling sites (Ranjani et al., 2021; Wang et al., 2021a). PLI is the pollution load index of MPs in a single site, n is the number of stations, and PLI_{zone} is defined as the pollution load index of MPs in the region.

2.6. Data analysis

The abundance unit of MPs is items·kg^{-1} d.w., and all data are expressed as the mean ± standard deviation (mean ± SD). The spatial and temporal differences in MP abundance were evaluated by one-way ANOVA, and the differences in the shape, color and particle size of MPs were analyzed by the Kruskal–Wallis test. All data were analyzed by SPSS V.26 statistical software, and the spatial distribution map of MP pollution was made by ESRI ArcGIS 10.2 software.

3. Results

3.1. Quality assurance and quality control (QA/QC)

In the environmental blank, the average abundance of MPs was 5.0 ± 12.4 items·kg^{-1} d.w., and this result was applied to the correction of microplastic abundance. At the same time, no MPs were detected in the blank membrane of the procedural background, indicating that there was no pollution from the program operation. Therefore, there was no need for blank correction.

3.2. Abundance of MPs

MPs were detected in all sampling sites in mariculture ponds and offshore areas, and the abundance of each sampling site is shown in Table 1. The average abundance of MPs in the mariculture area and offshore area was 49.2 ± 35.9 items·kg^{-1} d.w. and 17.1 ± 9.9 items·kg^{-1} d.w., respectively. In summer, the range of MP abundance in mariculture ponds was 14.2–172.6 items·kg^{-1} d.w., and the average was 59.3 ± 46.0 items·kg^{-1} d.w. However, in summer, the MP abundance range in offshore areas was 4.6–38.2 items·kg^{-1} d.w. Meanwhile, in winter, the range of MP abundance in mariculture ponds was 14.2–67.0 items·kg^{-1} d.w., and the average was 39.2 ± 19.4 items·kg^{-1} d.w. However, in winter, the MP abundance range in offshore areas was 4.6–38.2 items·kg^{-1} d.w. with an average of 17.6 ± 9.6 items·kg^{-1} d.w. The highest MP abundance occurred in the summer mariculture area, which was 172.6 items·kg^{-1} d.w., while the lowest MP abundance was 4.6 items·kg^{-1} d.w., which was in both summer and winter offshore areas. At the

Table 1
Microplastic abundance at each sampling site in sediments collected from Qingduizi Bay (items·kg^{-1} d.w.)

Aquaculture pond				Offshore area			
Summer		Winter		Summer		Winter	
Location	Abundance	Location	Abundance	Location	Abundance	Location	Abundance
P1	172.6	P1	52.6	S1	9.4	S1	14.2
P2	71.8	P2	67.0	S2	9.4	S2	4.6
P3	52.6	P3	19.0	S3	14.2	S3	9.4
P4	14.2	P4	43.0	S4	38.2	S4	14.2
P5	28.6	P5	67.0	S5	19.0	S5	9.4
P6	33.4	P6	28.6	S6	28.6	S6	23.8
P7	47.8	P7	14.2	S7	9.4	S7	23.8
P8	38.2	P8	33.4	S8	23.8	S8	38.2
P9	95.8	P9	47.8	S9	4.6	S9	19.0
P10	38.2		19.0	S10	9.4	S10	19.0
Average	59.3		39.2		16.6		17.6
Standard deviation	46.0		19.4		10.7		9.6
Mean ± standard deviation		49.2 ± 35.9		17.1 ± 9.9			

same time, we also found that the abundance of MPs in the estuary area (P1, P2, P9, P10) was higher than that in other areas.

3.3. Spatiotemporal distribution of MPs

The average abundance of MPs in the mariculture ponds (49.2 ± 35.9 items·kg^{-1} d.w.) was 2.8 times higher than that in the offshore area (17.1 ± 9.9 items·kg^{-1} d.w.). The abundance of microplastics in the sediments of mariculture ponds was significantly higher than that in offshore waters (ANOVA, $P < 0.05$), which indicated that mariculture caused microplastic pollution to some extent. Moreover, the coefficient of variation of MP abundance in mariculture ponds (0.72) was also larger than that in offshore areas (0.58). The MP abundance of estuary site P1 (172.6 items·kg^{-1} d.w.) in mariculture ponds was much higher than that of other sites. In the mariculture area, the closer the sea was, the less MPs abundance was found in the sediment. However, the MPs were evenly distributed in sediments of the offshore area.

There was no significant difference in MP abundance in sediment in either mariculture ponds or offshore areas between summer and winter (ANOVA, $P > 0.05$). The MP abundance in this area did not change with seasonal change. Moreover, our results showed that the shape composition of MPs had no seasonal change, because the MPs were mainly fiber in summer (99.44%) and winter (97.79%). Furthermore, the color and particle size of MPs had nothing to do with the season (Kruskal - Wallis, $P > 0.05$).

3.4. Morphological characteristics of MPs

3.4.1. MPs shape

In previous studies, MPs could be divided into four forms according to their morphology: fiber, pellet, fragment, and film (Zhao et al., 2018; Zhang et al., 2019). However, in this study, only fibers and fragments were found in the sediments (Fig. 2). Overall, there was no significant difference in the shape distribution of microplastics (ANOVA, $P > 0.05$). The shape of MPs in the mariculture ponds was mainly fiber (98.67%), and fragments were found only at 3 sites, accounting for 1.33% of collected MPs, whereas the MPs in the offshore areas were mainly composed of fibers (98.88%) and fragments (1.12%) (Fig. 3a). In summer, fibers accounted for 99.44% and fragments accounted for 0.56% of collected MPs. In winter, fibers accounted for 97.79%, and fragments accounted for 2.21% of collected MPs.

3.4.2. MPs color

MPs in this study area mainly included transparent, black, blue, red, and yellow colors. The main colors of MPs in the mariculture ponds were transparent (54.42%) and black (28.32%), while blue (4.87%), red (6.19%), yellow (2.21%) and others (3.98%) each accounted for less than 10%. The main color of MPs in offshore areas was also transparent, accounting for 69.66%, followed by black (12.36%), red (8.99%), blue (6.74%), and yellow (2.25%) (Fig. 3b). Overall, there were significant differences in the color of microplastics (ANOVA, $P < 0.05$). The variety of MP colors in the mariculture ponds was greater than that in the offshore area; in the mariculture ponds, the transparent MPs were nearly

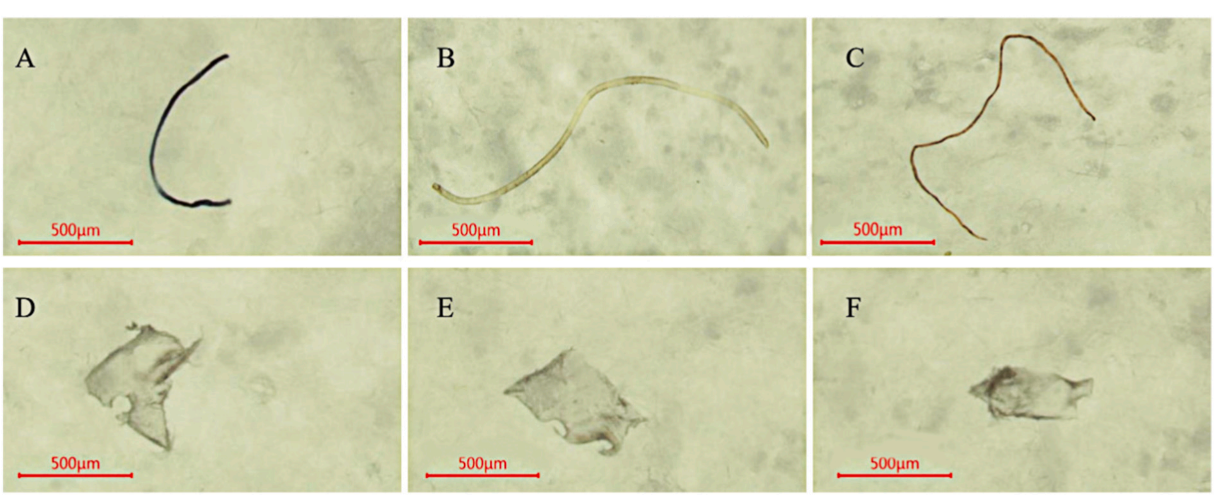

Fig. 2. Images of typical microplastics (Fibers: A–C; Fragments: D–F).

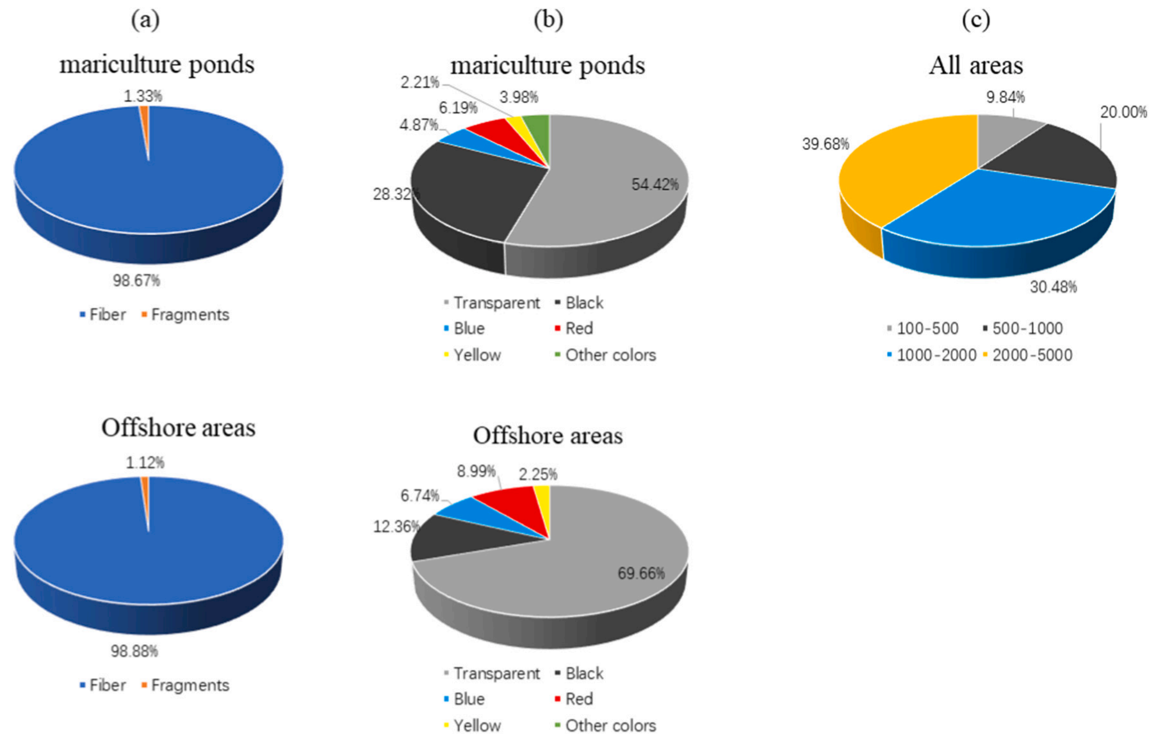

Fig. 3. The shape (a), color (b) and size distribution (c) of microplastics in sediments collected from Qingduizi Bay.

equal in percentage to colored MPs; on the other hand, in the offshore area, the percentage of transparent MPs was twice as much as the percentage of colored MPs. No obvious seasonal difference in MP colors was observed. The main colors of MPs in summer and winter sediments were both transparent (54.19% and 64.71%, respectively) and black (28.49% and 17.65%).

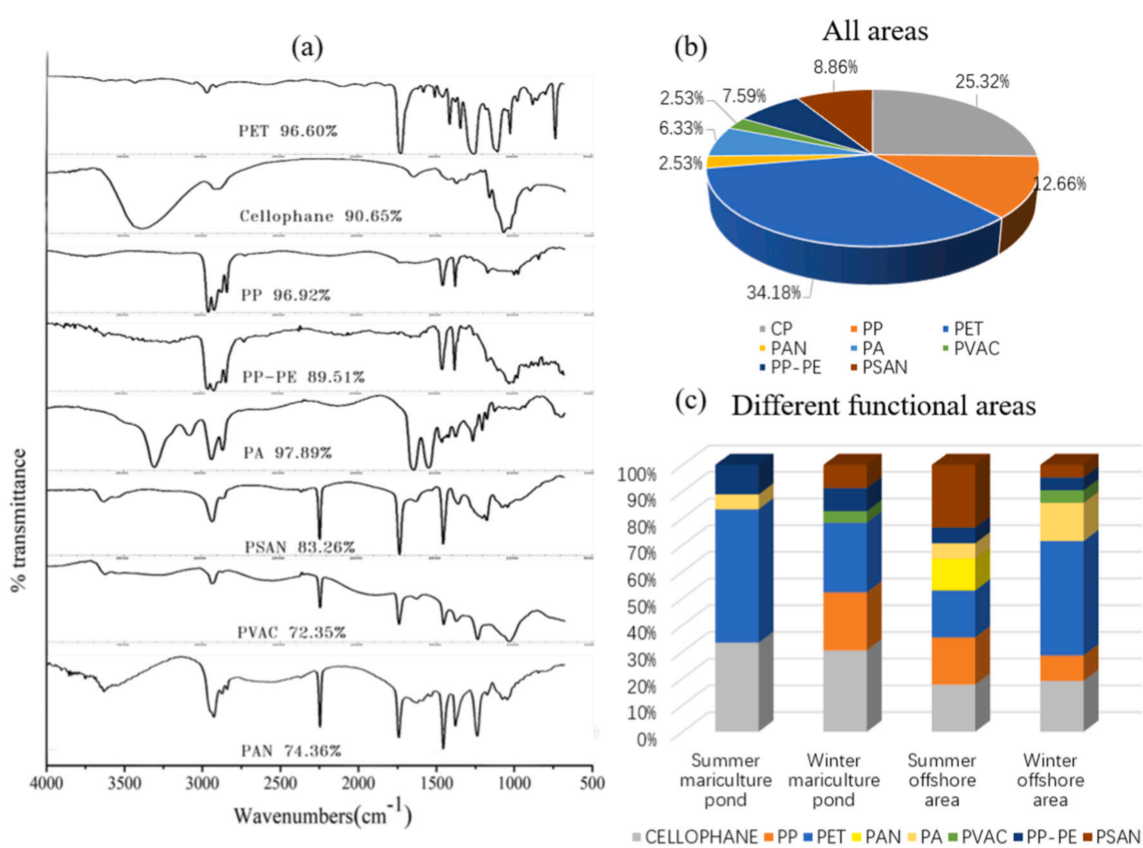

Fig. 4. Infrared spectra (a) and composition distribution diagram (b, c) of microplastics in sediments collected from Qingduizi Bay (b: Percentage of all areas; c: Percentage of different functional areas).

3.4.3. MPs size

MPs were divided into four size ranges, 100–500 μm, 500–1000 μm, 1000–2000 μm and 2000–5000 μm, as shown in Fig. 3c. The most common size range was 2000–5000 μm (39.68%), followed by 1000–2000 μm (30.48%), 500–1000 μm (20.00%), and 100–500 μm (9.84%). Overall, there was a significant difference in the particle size of microplastics (ANOVA, $P < 0.05$), and the distribution of different particle sizes was uneven. In this study area, as the size of MPs decreased, the proportion of MPs decreased to a certain extent. The minimum size observed in the whole region was 117.02 μm, which was observed at Station P3 in the summer mariculture ponds.

3.4.4. MPs polymer

A total of 109 suspected MPs were identified by μ-FTIR, and the results were compared with the spectral database of OMNIC 8.2 software; 79 samples were identified as MPs, with a success identification rate of 72% (the matching degree was more than 70%) (Teng et al., 2019). Eight different types of polymers were identified in all samples, and their infrared spectra are shown in Fig. 4a. The main types include polyethylene terephthalate (PET, 34.18%), cellophane (CP, 25.32%), polypropylene (PP, 12.66%), poly(styrene:acrylonitrile) (PSAN, 8.86%), polypropylene-polyethylene copolymer (PP-PE, 7.59%), polyamide (PA, 6.33%), polyvinyl acetate (PVAC, 2.53%), and polyacrylonitrile (PAN, 2.53%). Among these, PET (34.18%) was the most common component, followed by CP (25.32%) (Fig. 4b, c). PET MPs were dominant in mariculture ponds and offshore areas, accounting for 36.59% and 31.58%, respectively, of collected MPs. There were more polymeric types of MPs in offshore areas than in mariculture ponds.

3.5. Risk assessment of MP pollution

The PLI value of each sampling point was calculated, and the results are shown in Table 3 and Fig. 5. According to the classification of pollution degree, all sampling sites were at Hazard Level I. The results showed that the MP pollution level of the sediments in Qingduizi Bay was low. The pollution level order was as follows: summer mariculture pond > winter mariculture pond > winter offshore area > summer offshore area (3.22 > 2.74 > 1.82 > 1.73). Moreover, the PLI value of MPs did not change significantly in summer and winter in this region. The PLI value of MPs in the mariculture pond was higher than that in the offshore area, indicating a higher risk in the mariculture pond area.

4. Discussion

4.1. Abundance of MPs in mariculture areas

As one of the main sources of MPs in the ocean, MPs in mariculture have attracted increasing attention in recent years (Dong et al., 2021). In this study, MPs were found in all sampling sites (mariculture ponds: 49.2 ± 35.9 items·kg^{-1} d.w., offshore area: 17.1 ± 9.9 items·kg^{-1} d.w.), indicating that there was MP pollution in Qingduizi Bay. Table 2 summarizes the research data on sediments and water bodies in mariculture areas, indicating widespread MP pollution in mariculture areas. It was reported that the average abundance of MPs in the sediments of mariculture farms in Xiangshan Bay was 74 items/kg, and the study showed that mariculture activities may increase the content of MPs in sediments (Wu et al., 2020a). The MPs in sediments from finfish mariculture in Spain ranged from 0 to 213 particles/kg (Kruger et al., 2020). The range of MPs found in the sediments of sea cucumber farms in the Yellow Sea and Bohai Sea was 1–52 items/50 g (Mohsen et al., 2019). Our results showed that the average MP abundance in Qingduizi Bay was at a moderate level compared with that found in other studies. However, our samples were collected in 2012, which was much earlier than other studies. Additionally, MPs were found in water bodies in both freshwater culture and mariculture areas, such as Honghu culture ponds (87–750 items/m^3) (Xiong et al., 2021) and Sanggou Bay mariculture water bodies (63.6 ± 37.4 items/L) (Wang et al., 2019a). However, the comparison between studies was hindered by methodological differences, such as sampling methods and units of measurement. Until recently, standardized sampling and separation methods for MPs have not been established worldwide (Stock et al., 2019).

4.2. The source and migration of MPs in mariculture areas

Mariculture has developed rapidly since 1991 in China, and mariculture products accounted for 76.5% of the total aquatic products in 2018 (FAO, 2020). Plastic products are widely used in the mariculture industry, which diversifies the sources of microplastics. The sources of MPs in mariculture areas can be divided into two parts: direct sources and indirect sources. The direct source was the intentional production of small plastics, such as feed and boat paint, and the indirect source was the decomposition of large plastics, such as fishing gear and plastic garbage. Fig. 3 and Fig. 4 depict the distribution of the shape, color, size and polymer types of MPs in the sediments of Qingduizi Bay. From these characteristics, we can further infer their origin (Shim et al., 2018). Fiber was the most abundant form of MP in this region, which was consistent with most studies on mariculture areas. It was speculated that fiber mainly came from fishing gear and textiles. Plastic fishing gear,

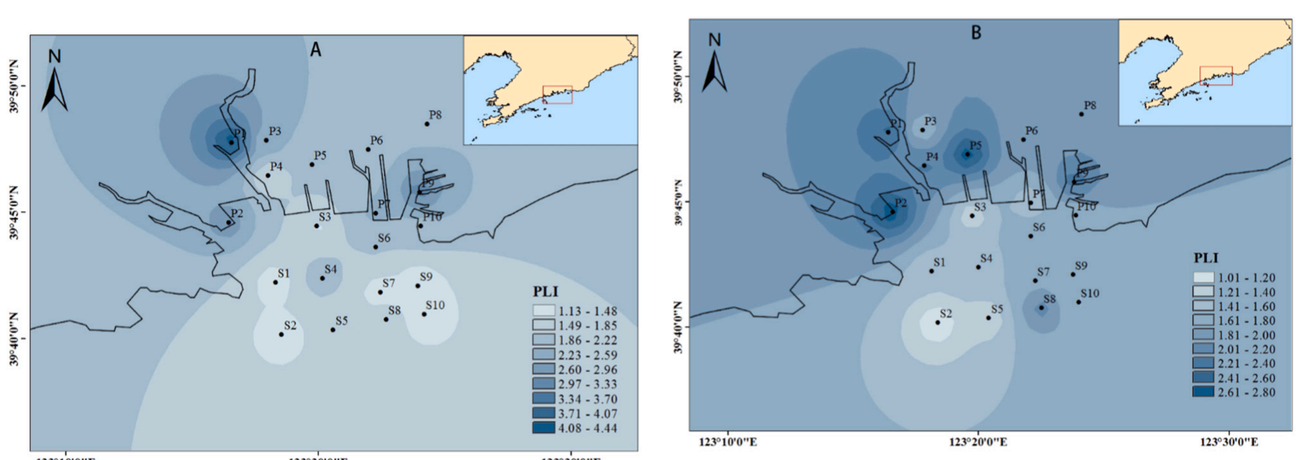

Fig. 5. Risk distribution map of microplastics (A: summer, B: winter).

Table 2
Comparison of microplastic abundance in different aquaculture areas.

Area	Source	Abundance	Shape	Composition	Reference
Aquaculture ponds in Honghu Lake, China	Freshwater	87–750 items/m^3	Fiber and fragment	PP, PET, PE	(Xiong et al., 2021)
Fish ponds in the Pearl River, China	Freshwater	42.1 particles/L	Fiber	PP, PE	(Ma et al., 2020)
Shrimp-culturing farm in Longjiao Bay, China	Seawater	250–5150 particles/m^3	Granule and fiber	PE, PET	(Chen et al., 2020)
Sanggou Bay, China	Seawater	63.6 ± 37.4 items/L	Fiber	PE, PP	(Wang et al., 2019a)
Shellfish farm in Jinhae Bay, Korea	Sediment	0.94 ± 0.69 particles/g	Fragments	PS	(Jang et al., 2020)
Jiaozhou Bay, China	Sediment	15 ± 6 items/kg	Fiber	PET	(Zheng et al., 2019)
Xiangshan Bay, China	Sediment	74 items/kg	Fiber	Cellulose, PP	(Wu et al., 2020a)
Fish farms in Western Mediterranean, Spain	Sediment	0–213 particles/kg	Fiber	PP, PE	(Kruger et al., 2020)
Sea cucumber farms in Yellow Sea and Bohai Sea, China	Sediment	1–52 items/50 g	Microfiber	Cellophane	(Mohsen et al., 2019)
Aquaculture pond in Qingduizi Bay, China	Sediment	49.2 ± 35.9 items/kg	Fiber	PET	Present study
Offshore area in Qingduizi Bay, China	Sediment	17.1 ± 9.90 items/kg	Fiber	PET	Present study

Table 3
Pollution grade level of microplastics and PLI value of microplastics in sediments collected from Qingduizi Bay.

The pollution load index (PLI)	<10	10–20	20–30	>30
Pollution grade level	I	II	III	IV
Summer aquaculture pond	3.22			
Summer offshore area	1.73			
Winter aquaculture pond	2.74			
Winter offshore area	1.82			

such as fishing lines, nets, and cages, is widely used in mariculture. Due to the erosion and aging caused by seawater and the destruction caused by ocean currents, monsoons and other dynamic factors, and due to some human activities such as trawling, the content of fiber MPs in marine fishery waters usually increases over time (Chen et al., 2021b). A study showed that there was a strong positive correlation (Pearson, $r = 0.9176$, $P < 0.05$) between fishing activity and microplastic abundance (Dowarah and Devipriya, 2019). Textiles were also a major source of microplastic fibers. Data have shown that more than 90 million metric tons of textile fibers were produced globally in 2016 (Gasperi et al., 2018), and improper handling of these textiles can lead to serious fiber microplastic pollution. Another reason for the high proportion of fibers in sediments might be that the polymers that make up the fibers were generally denser and settled more easily in seawater (Maes et al., 2017). As far as color was concerned, transparent, black, blue, yellow, red and other colors were observed, and it was speculated that they might come from human activities, such as fishing or tourism. James et al. (2021) pointed out that intensive tourism and fishing activities can cause microplastic pollution. Additionally, it was also observed that some of the fibers had faded blue and white colors, which might be due to the release of some dyes from aging cages and ropes in many mariculture farms (Zhang et al., 2021a).

The size of MPs was largest in the range of 2000–5000 μm, which further verified that the MPs in this area were mainly broken and decomposed from large plastics. A total of seven kinds of MP components were found in this region, indicating that the MP sources in the region were diverse. The main polymer type was PET, which was consistent with the study in Jiaozhou Bay and Laizhou Bay (Zheng et al., 2019; Teng et al., 2020). It was reported that 6 million MP fibers could be released from a 5 kg load of PET textiles (Sait et al., 2021). Moreover, PET had a high density (1.37–1.45 g/cm^3) and easily settled (Bellasi et al., 2021). In recent years, CP, which is an organic cellulose-based polymer (Teng et al., 2020), has also been considered a kind of microplastic (UNEP, 2016; Nurhasanah et al., 2021; Yuan et al., 2021; Gong et al., 2021). At the same time, it is also a typical semisynthetic material, to which a large number of additives are usually added. CP is often used in food packaging bags (Su et al., 2016). This microplastic type was found in many regional sediments. Studies have shown that CP was the most abundant polymer type in Ma'an Archipelago deposits (47.8%) (Zhang et al., 2020), and CP accounted for 16.6% of polymers in Hangzhou Bay (Li et al., 2020b). Furthermore, feed and fertilizer in mariculture are also important sources of MPs (Lv et al., 2020).

As mariculture ponds are generally located in semienclosed bays, the fluidity of seawater is relatively small (Chen et al., 2018). Therefore, they can be regarded as potential storage pool of MPs that accumulate MPs during mariculture. These areas could also be sources of MPs that spread to offshore areas and further migrate in the ocean due to the flow of seawater or human disturbances.

4.3. Spatiotemporal variation in MPs in sediments of mariculture areas

Our research showed that there was no significant temporal change in the abundance and distribution characteristics of MPs in sediments, which was similar to the results of the study of Sanggou Bay (Sui et al., 2020b). Our results suggested that most of the MPs in sediments accumulated for a long time. This observation might also be related to the geographical location of Qingduizi Bay. Since it is a semiclosed bay, the exchange rate between mariculture and external water was relatively low, so it easily accumulated MPs (Xue et al., 2020). Furthermore, the MPs in sediments were less affected by rainfall and river water flow; therefore, no obvious seasonal change in MPs was observed in sediments of this area. However, several studies have shown that the distribution of MPs varies seasonally. Chen et al. (2021a) found that MP abundance in the dry season was higher than that in the rainy season (rainfall) in surface sediments along the southwest coast of Taiwan. The concentrated rainfall and substantial runoff in the rainy season can wash land MPs into the sea, resulting in low MP abundance (James et al., 2021). However, the precipitation in our study area was relatively small in summer.

The distribution of MPs in sediment showed significant spatial changes in this region, and the factors that affect the distribution of MPs mainly include human activities, emission sources, plastic characteristics, and meteorological and hydrodynamic conditions (Qi et al., 2020). There was a significant difference in the abundance of MPs between the mariculture ponds and the offshore area. The high abundance of MPs in the mariculture ponds might came from the large number of plastic mariculture facilities, which were broken into MPs because of aging, weathering and abandonment. The low MP abundance in the offshore area might be due to seawater dilution or migration by external forces (wind, waves, tides). The spatial distribution of MPs in the sediments of Qingduizi Bay showed a downward trend from the inside to the outside, which was similar to the distribution of MPs in sediments of Sanggou Bay (Sui et al., 2020b) and Xiangshan Bay (Chen et al., 2018). The main reason for this distribution trend is that there are a large number of mariculture activities in the bay, as well as abundant human activities and the inflow of urban wastewater. The water in the bay is less mobile, which is likely to contribute to the deposition of microplastics. However, the close proximity to the sea implies less microplastics due to the dilution of seawater. Moreover, at some sites (P1, P9) near the river entrance, the abundance of MPs was higher than at others, possibly due to wastewater from human activities, such as laundry wastewater. The MP abundance was high at site P1 in the mariculture pond because it was located near the Huli River estuary, and it received many land-sourced MPs from the river. Another high MP abundance site was detected at P9 because there were more towns and more frequent human activities around this area. Zheng et al. (2020) also proved that the abundance of MPs was higher near estuaries.

4.4. Risk assessment of MP pollution in mariculture area

Edible aquatic products are an important source of protein in the human food supply. Mariculture products contaminated by MPs will directly affect the quality and safety of food. Widespread MP pollution in cultured seafood has aroused concern about human health (Amaral-Zettler et al., 2020). According to a hazard-ranking model based on the United Nations' Globally Harmonized System of Classification and Labelling of Chemicals, chemical ingredients that are >50% plastics are hazardous (Ranjani et al., 2021). At present, MPs have been found in many commercial aquatic products. Examples of contaminated products include 10 species of commercial marine fish in the Bay of Bengal (Ghosh et al., 2021), 29 species of commercial fish in the Bohai Sea (Wang et al., 2021b), 24 species of fish in marine farms in the East China Sea (Wu et al., 2020b), and the cultured Zhikong scallop *Chlamys farreri* (Sui et al., 2020a). The level of MPs in these aquatic products might be affected by environmental variables such as the sediments, so the pollution risk of MPs in sediments was evaluated by the pollution load index (PLI). Mai et al. (2021) used PLI to assess the risk of MPs in the Xijiang River and Pearl River Estuary, and the results showed that the risk was relatively low. Liu et al. (2020) showed that the risk of MP pollution in Jiaozhou Bay was moderate ($PLI_{JZB} = 11.76$). The pollution risk assessment of MPs in the Dongshan Bay Estuary was at Hazard Level II ($PLI = 14.2$) (Pan et al., 2021). The PLI values of the Yangtze Estuary and the adjacent East China Sea were 18.4 and 20.4, respectively (Xu et al., 2018). Li et al. (2021b) used the PLI to evaluate the pollution risk of MPs along the coast of Guangdong, and the overall risk level was moderate. The current pollution level in this region was slight, which might be related to the early sampling time. The MP pollution level was bound to increase with increasing mariculture activity intensity, so it was necessary to take preventive measures in advance. However, the risk assessment presented here has some limitations. In the future, there is an urgent need to develop a unified research method of MPs and to conduct more extensive research on the potential pollution risks brought by MPs.

5. Conclusion

MPs were found in the sediments of the mariculture area and the adjacent sea area in Qingduizi Bay. The abundance of MPs in sediments of the mariculture pond and offshore area was 49.2 ± 35.9 items·kg^{-1}d.w. and 17.1 ± 9.9 items·kg^{-1}d.w., respectively. The MPs collected in this study were mainly transparent fibers with a size of 2000–5000 μm. Seven different types of polymers were identified, of which PET and cellophane were the main components. There were significant differences in MP abundance between mariculture areas and offshore areas. There was a downward trend of abundance from the inside to the outside. This study showed that mariculture activities may increase the content of MPs in sediments. The pollution load index (PLI) risk assessment showed that all sampling sites were at Hazard Level I, indicating that the risk of MP pollution in the sediments of Qingduizi Bay was low. However, measures should be taken to reduce the release of MPs in mariculture areas.

CRediT authorship contribution statement

Liang Chen: Investigation, Software, Formal analysis, Writing – original draft. **Xiutang Yuan:** Writing – original draft, Methodology. **Yuheng Ye:** Methodology, Investigation. **Jia Teng:** Software, Methodology. **Jianmin Zhao:** Writing – review & editing, Funding acquisition. **Qing Wang:** Conceptualization, Supervision, Writing – review & editing, Funding acquisition. **Bin Zhang:** Conceptualization, Methodology, Funding acquisition.

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.

Acknowledgments

This study was supported by the National Key Research and Development Project (2019YFD0901101), the Strategic Priority Research Program of the Chinese Academy of Sciences (No. XDA23050303), the National Natural Science Fund of China (42176161, 41576122), the Two-Hundred Talents Plan of Yantai (Y839081021), the Open Research Fund of State Key Laboratory of Estuarine and Coastal Research (SKLEC-KF201908), the Project of State Key Laboratory of Environmental Chemistry and Ecotoxicology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences (No. KF2020-16), the Ministry of Education Chun Hui Plan Project (No. 191650).

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