Environmental Pollution 273 (2021) 116450

Contents lists available at ScienceDirect

Environmental Pollution

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

Long-term trends of microplastics in seawater and farmed oysters in the Maowei Sea, China☆

Jingmin Zhu [a], Qiang Zhang [a], Yinan Huang [a], Yuping Jiang [a], Jiana Li [d], Jennifer J. Michal [e], Zhihua Jiang [e], Youhou Xu [a], Wenlu Lan [a, b, c, *]

[a] Guangxi Key Laboratory of Marine Disaster in the Beibu Gulf, Ocean College, Beibu Gulf University, Qinzhou, 535011, China
[b] Guangxi Academy of Sciences, Guangxi Mangrove Research Center, Guangxi Key Lab of Mangrove Conservation and Utilization, Beihai, 536000, China
[c] Marine Environmental Monitoring Center of Guangxi, BeiHai, 536000, China
[d] College of Oceanography, Hohai University, Nanjing, 210098, China
[e] Department of Animal Sciences and Center for Reproductive Biology, Washington State University, Pullman, WA, 99164-7620, USA

ARTICLE INFO

Article history:
Received 13 July 2020
Received in revised form
11 December 2020
Accepted 5 January 2021
Available online 8 January 2021

Keywords:
Microplastics
Oysters
Maowei Sea
Time trends
Accumulation

ABSTRACT

Microplastic pollution in marine environments and organisms has received a great deal of international attention. However, the long-term field studies of microplastics are rare. Here, we assessed annual variation in microplastic abundance in the Maowei Sea, a classic mariculture bay in southern China, and analyzed the long-term accumulation in oyster tissues. U-shaped time trends of microplastics in water were observed from January to December in 2018 in the estuarine region, inner bay, and mouth bay sites, representing an inverse relationship with the local rainfall patterns. The common microplastic particles in Maowei Sea are PET/PE fibers, and polystyrene foams, which are mainly related to textile pollution and fishery activities. After one year of continuous monitoring, we did not find accumulation of microplastics in the whole soft tissues of oyster after 10% KOH digestion. No significant correlation of microplastic abundances between water and oysters was observed. The microplastic abundance in oyster was correlated with some environmental variables (i.e. salinity, pH, nutrients and total organic carbon) of the surrounding water following Spearman correlation analysis. The microplastic levels in oysters could probably be influenced by the environmental variables.

1. Introduction

Microplastics are omnipresent in marine systems globally. Plastic particles with sizes ranges from 1 μm to 5 mm in diameter are generally considered microplastics (Frias and Nash, 2019; GESAMP, 2015). Microplastics are of concern because the small particles can be ingested by a variety of organisms and eliminating them from the environment is difficult (Rocha-Santos and Duarte, 2015; Hollman et al., 2013; Chen et al., 2019; Hurley et al., 2017). Microplastic abundance in estuarine ecosystems has been reported to be much higher than in surrounding ecosystems (Zhang, 2017; Fok and Cheung, 2015). For example, Hong Kong, located at the Pearl River Estuary, is a microplastic pollution hotspot (Fok and Cheung, 2015). Estuaries provide habitats for a high number of organisms and have high productivity levels (McLusky and Elliott, 2004), and support many fishery and mariculture bays. Abandoned, lost, or otherwise discarded fishing gears are thought to be the main contributors to the generation of microplastics in aquatic environments in the fishery and mariculture sectors (Lusher et al., 2017). The commercial gears which have the potential for greatest contribution to microplastic loads are the nettings used in benthic dredges and trawls and in particular the ground ropes (Lusher et al., 2017). In addition to the in-use degradation of fishing gears, and despite careful maintenance, analysis of the abundance of abandoned, lost or otherwise discarded fishing gears have shown that many kilometres of netting are lost to the marine environment each year (Macfadyen et al., 2009). Currently, only a handful of studies have sought to quantify microplastics in mariculture bays, and the reports are restricted to China (Chen et al., 2018; Feng et al., 2019; Zhu et al., 2019) and Brazil (Castro et al., 2016).

A mariculture bay is an enclosed area of the ocean, with multiple in-flowing rivers or streams, and a link to the open sea. As the

☆ This paper has been recommended for acceptance by Maria Cristina Fossi.

* Corresponding author. Guangxi Key Laboratory of Marine Disaster in the Beibu Gulf, Ocean college, Beibu Gulf University, Qinzhou, 535011, China.
E-mail address: dr.lan@139.com (W. Lan).

https://doi.org/10.1016/j.envpol.2021.116450
0269-7491/

transition zones between river and maritime ecosystems, mariculture bays receive land, river, and sea-based microplastic inputs, including surface runoff, sewage discharge, effluents from industrial plants, tourism, riverine discharge, shipping, fisheries, and mariculture (Auta et al., 2017; Zhang, 2017; Lusher et al., 2017; Browne et al., 2011; Wagner et al., 2014). Microplastic monitoring in mariculture bays is essential for elucidating sources, transport, and distribution of microplastics. Furthermore, mariculture is an important source of seafood globally, and seafood has been the most studied dietary intake route of microplastics (Lusher et al., 2017; Cox et al., 2019). In general, fish species are consumed after removing their gastrointestinal tracts, while most bivalves are consumed whole, which greatly increases the risk of microplastic ingestion. However, most publications on the contamination of microplastics in bivalves mainly refer to wild sources, with less attention on farmed sources (Lusher et al., 2017).

The ingestion of microplastics by aquatic organisms, particularly commercially important species, has increasingly attracted the attention of researchers. Mussels and oysters are used extensively in laboratory exposure experiments to investigate the uptake, accumulation, and clearance of microplastics. Bioaccumulation is a dynamic process involving contaminants in the environments and their uptake by exposed organisms through time, and then toxicity can manifest after bioaccumulation occurs (Wang, 2016). Filtration rate and particle size of microplastics have been reported as important factors in the biodynamics of microplastics in bivalves (Setälä et al., 2014; Van Cauwenberghe et al., 2015). Filtration rate of bivalves is regulated by the nutrient levels in the water and growth status of bivalves (Gosling, 2008). It is remains unclear if bivalves bioaccumulate plastic particles. Qu et al. (2018) reported a significant linear increase in microplastic ingestion in mussels after 5 days of increasing exposure. Conversely, related laboratory experiments suggest bivalves do not bioaccumulate microplastics (Fernández and Albentosa, 2019; Rist et al., 2019; Xu et al., 2017). Notably, organisms in most laboratory researches are exposed to unrealistically high doses of microplastics over relatively short exposure times (Lambert et al., 2017; Koelmans et al., 2017; Rochman et al., 2016). Only a few field studies have investigated the accumulation of microplastics by aquatic organisms based on correlation with field data (Li et al., 2018a,b; Su et al., 2016). There is a lack of long-term field reports on the accumulation of microplastics during growth in organisms.

Microplastics are distributed unevenly in water bodies. However, most studies obtained samples only one or two times during the dry and wet seasons (Eo et al., 2019; Rodrigues et al., 2018; Frere et al., 2017; Lima et al., 2014; Cheung et al., 2018). Eo et al. (2019) demonstrated seasonal variation in microplastic abundance in the Nakdong River, South Korea, reporting that 70–80% of the annual microplastic load occurred in the wet season. The findings are consistent with observation made from the eight major rivers of the Pearl River Delta, China, with the winter and summer seasons having the lowest and highest riverine inputs, respectively (Mai et al., 2019). Such variations can be explained by the difference in water discharges. It is necessary to monitor at least between the wet and dry seasons to acquire the representative level of microplastic pollution (Shim et al., 2018). Currently, there are significant knowledge gaps on how microplastics vary with time and across seasons.

We assessed the long-term trends in microplastic concentrations in a mariculture zone and the correlation between field concentrations and accumulation by bivalves in the Maowei Sea, a semi-closed area in the northwestern Beibu Gulf, South China Sea. We found in a previous study that microplastics are universal in the Maowei Sea (Zhu et al., 2019). In the present study, microplastics with sizes >5 μm in diameter were sampled monthly from January–December 2018 from surface water and oysters in estuarine region, inner bay, mouth bay, and open bay sites of the Maowei Sea. Environmental variables including water temperature, pH, salinity, dissolved oxygen, total organic carbon, total nitrogen, total phosphorus, chlorophyll *a*, and zooplankton biomass were also measured. Furthermore, potential correlations and factors influencing long-term microplastic trends in surface water and oysters were analyzed.

2. Material and methods

2.1. Sample collection

Sampling sites were selected in the estuarine region (S1), inner bay (S2), mouth bay (S3), and open bay (S4) locations of Maowei Sea (Fig. 1), the detailed information is presented in Table S1. The sites were located from north to south in the Maowei Sea, and the salinity in the sites ranged from 0 to 25 psμ. We rented oyster rafts from the local fishermen at each site, and the oysters were transplanted to the sites to be monitored from January to December 2018. At each site, three 5-L surface water (<10 cm in depth) were collected using a steel bucket, and the water were filled into glass bottles. A string of oysters (60–100 cm, about 10 individuals each) was collected from the farmed raft at each site. Surface water samples were collected every mid-month from January to December 2018, yielding 12 sampling events. Oyster samples were collected monthly beginning in the second month (February), resulting in a total of 11 sampling events. We failed to collect oyster samples at S4 in February because of the bad weather.

Environmental variables including water temperature, pH, salinity, dissolved oxygen, total organic carbon, total nitrogen, total phosphorus, chlorophyll *a*, and zooplankton biomass were also measured. Zooplankton samples were obtained by vertical hauls from about 1 m above the seafloor to the surface using a plankton net (505 μm mesh size, 50 cm mouth diameter), then preserved in a 1-L polyethylene bottle with formalin at a final concentration 5%. The volume of filtered water was obtained by the rope length multiplied with the mouth area (m^3). Water temperature, pH, salinity, and dissolved oxygen (DO) were measured *in situ* by YSI 6600 multi-probes sensor (USA). Water samples were collected using 5-L Polyethylene bottles at about 0.5 m below the surface of the seawater for total organic carbon, nutrients, and chlorophyll *a* analysis.

2.2. Quality control of the experiments

To avoid microplastic contamination, the following methods described in our previous study (Zhu et al., 2019) were employed in this study. Cotton clothes were worn during all steps of sampling and analytical procedures. All liquids, such as potassium hydroxide (KOH) and tap water, were filtered through a 1-μm-pore size Nylon membrane before use. All lab supplies were rinsed with filtered pure water three times. The samples were capped with lids or covered with tinfoil promptly after every operation. Three blanks were prepared using three glass bottles, and the lids remained open during sampling. The field blanks were transported back to the laboratory and analyzed along with the field samples. In the laboratory, blanks were filled with filtered pure water, and were treated with the same protocol used for samples. The reported abundances were corrected using average blank concentration.

2.3. Isolation of microplastics

The protocol for extraction of microplastics from water followed

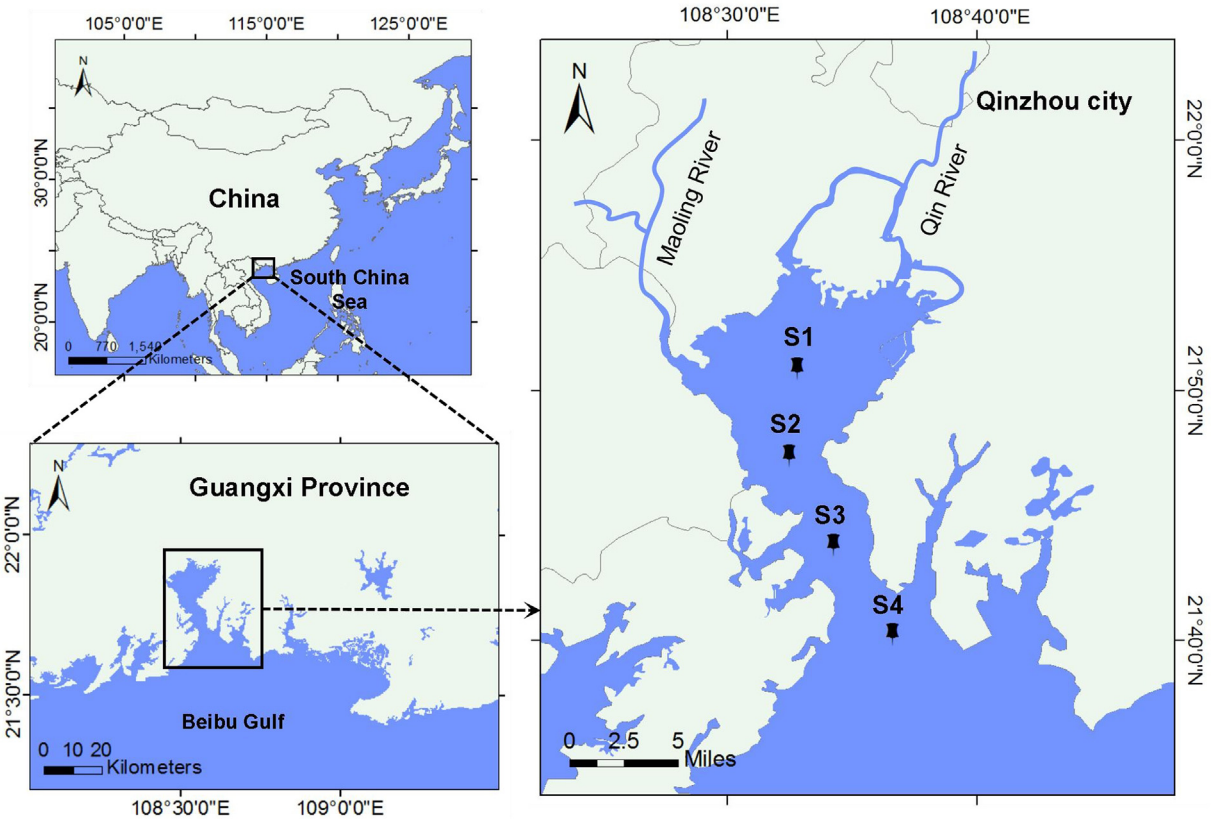

Fig. 1. Geographical locations of long-term sampling sites in Maowei Sea.

procedures of Gago et al. (2018) with some modifications. Water samples were filtered onto a Nylon membrane with 47 mm diameter and 5-μm-pore size (Millipore, Burlington, MA, USA, NY2004700) facilitated with a vacuum pump (FY-3C-N, VALUE, China). Next, materials on the membrane, including organic matter, were carefully washed into a 250 mL conical flask. Potassium hydroxide solution was added at 1:3 vol sample:solution ratio to digest the biological material (10% KOH was prepared and filtered through a 1-μm-pore size membrane). The mixture was placed in a vapour-bath constant temperature sample shaker (SHZ-82, Shanghai, China) at 40 °C and stirred at 80 rpm for about 48 h. The digested materials were pumped onto a Nylon membrane with 47 mm diameter and 5-μm-pore size (Millipore, NY2004700). After that, the membrane was stored in a clean Petri dish with lid for further examination.

A string of oysters was divided into top, center, and bottom groups depending on the depth. The tissues from the oysters in each group were pooled together during analysis, and the concentration per individual is the group concentration divided by three. Nine individuals were totally analyzed in one string of oysters. The weights and lengths of oysters were recorded, and then the gills, digestive glands, and other tissues were removed (Fig. 4A) and weighed (Table S2). The above tissues were digested following our previous study with minor modifications (Zhu et al., 2019). Briefly, soft tissues were digested with 10% KOH liquid at 40 °C with stirring at 80 rpm for about 48–72 h in a vapour-bath constant temperature sample shaker (SHZ-82, Shanghai, China). The digested materials were pumped onto a Nylon membrane with 47 mm diameter and 5-μm-pore size (Millipore, NY2004700). After that, the membrane was stored in a clean Petri dish with lid for further examination.

2.4. Observation and validation of microplastics

All filters were observed under a Cnoptec SZ680 stereomicroscope (Chongqing, China), with all images obtained using an AxioCam digital camera. All particles from each sample were transferred to a cellulose nitrate grid filter (Whatman WME, 47 mm diameter, 0.45-μm-pore size) for micro-Fourier transformed infrared spectroscopy (μ-FT-IR, Nicolet iN10 MX, Thermo Fisher Scientific, USA) identification in transmittance mode (Hu et al., 2018; Hermsen et al., 2018). The particles on the cellulose nitrate grid filters were transferred onto the microcompression cell II using a tweezer for identification with diamond window using FTIR instrument (Zhang et al., 2020). Considering that large amounts of fibers were predominant in all samples, 5–10 fiber particles from each sample were randomly chosen and identified, while other shape particles were all analyzed. In total, 764 fibrous and 902 non-fibrous particles were identified. The spectrum was compared straightway with the library of polymers supplied by the software of Thermo Fisher Scientific (OMNIC Picta). Spectra with quality index of >60% were accepted in this study (Yang et al., 2015). The reported characteristics of microplastics (i.e. color, shape and size) were corrected using their type-specific concentrations found in the blank. If there were no same type particles found in the samples, the corresponding type-specific concentrations remain unchanged. Because not all particles were identified using μ-FT-IR, the polymer characteristics was corrected using specific blank concentrations multiplied by μ-FT-IR identified ratio.

The physical properties of microplastics were identified based on Gago et al. (2018). The identified colors were white, yellow, blue, green, red, and black. The shape of microplastics was categorized by their morphological characteristics: fiber (Fig. 2A), film (Fig. 2B), foam (Fig. 2C) and fragment (Fig. 2D). The size of microplastic

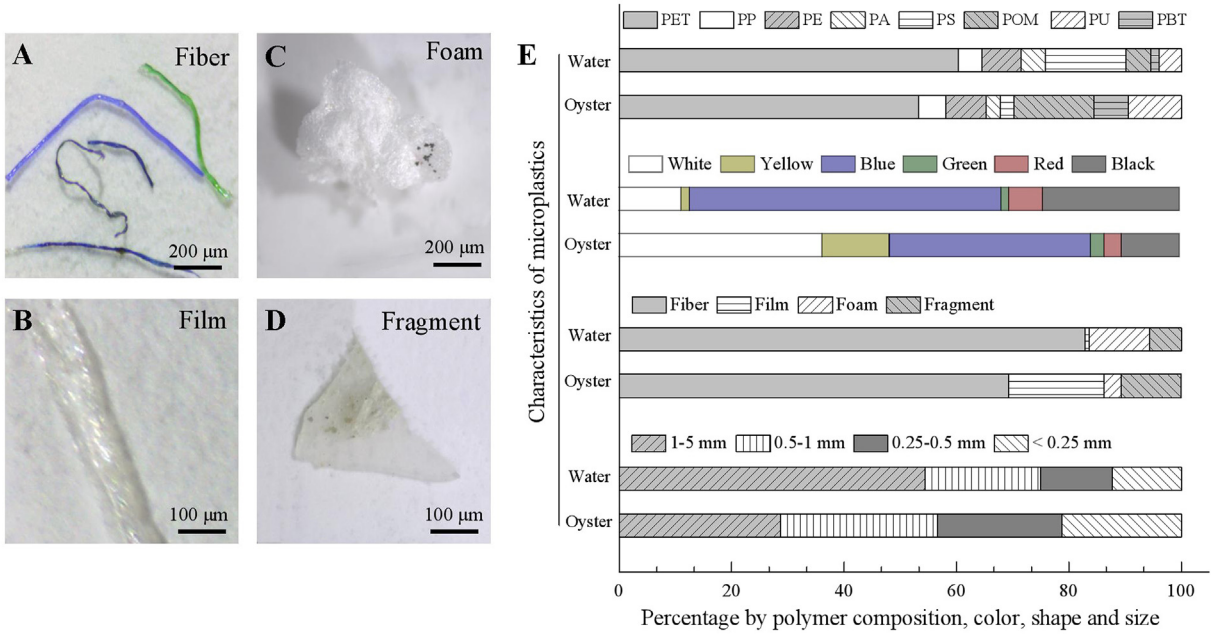

Fig. 2. Characteristics of microplastics in water and oyster samples. Abbreviations: PET: polyester; PP: polypropylene; PE: polyethylene; PA: polyamide (nylon); PS: polystyrene; POM: polyoxymethylene; PU: polyether urethane; PBT: polybutylene terephthalate.

particles was obtained by measuring the longest dimension with ImageView 3.7 software, and were classified into four groups: 1–5 mm, 0.5–1 mm, 0.25–0.5 mm, < 0.25 mm (Zhu et al., 2019).

2.5. Laboratory analysis of environmental variables

Total nitrogen and total phosphorus samples were pretreated using alkaline persulfate digestion and potassium persulphate digestion, respectively, then were determined using a AutoAnalyzer 3 (SEAL, Germany). For chlorophyll *a* (Chl-a), the water samples were filtered onto a 0.7 μm GF/F-filter (Whatman, England). The filters were evenly soaked with 1–2 drops of saturated magnesium carbonate. The Chl-a concentration was determined with an ultraviolet visible spectrophotometer (DR6000, HACH, USA) after extraction with 90% acetone (v/v) in darkness for 24 h at 4 °C. Total organic carbon concentration was directly determined using a TOC-L analyzer (Shimadzu, Japan). Zooplankton biomass were measured using weight measurement (Table S3).

2.6. Data analysis

The microplastic abundance in water was expressed as average particles per liter ± standard error of mean (SEM) (n = 3), using standard propagation of error methods (Bevington et al., 1993). The microplastic abundance in oyster samples (three replicates, nine individuals) was expressed as average particles per wet gram of digested tissue and per individual ± SEM respectively. Statistic analysis was performed using IBM SPSS Statistics (IBM Corp., Armonk, NY, USA), GraphPad Prism v5.0 (GraphPad Software Inc., San Diego, CA, USA), and Origin 9.0 (OriginLab Corporation, Northampton, MA, USA) software. Mean differences of microplastic abundances among samples were assessed using repeated measures ANOVA followed by Dunnett post-hoc test. Independent samples *t*-test was used for each two group comparison. The repeated measures ANOVA and *t*-test models were first checked for normality and homogeneity of variables. If the variables was not uniform, the dependent variable was log-transformed. The correlation of microplastic abundances and environmental variables of surrounding water was evaluated with Spearman correlation rank, using IBM SPSS Statistics software.

3. Results

3.1. Identification and characteristics of microplastics

Contamination of 0.7 ± 0.4 particles per blank was detected (n = 3), which may be caused by airborne microplastics (Dris et al., 2015; Zhang et al., 2020). The final abundance was corrected by the procedural blank data. The most abundant polymers in water samples were polyester (PET, 60.3%) followed by polystyrene (PS, 14.3%) and polyethylene (PE, 7.0%). In oyster samples, PET (53.2%) also dominated but were followed by polyoxymethylene (POM, 14.2%) and polyether urethane (PU, 9.5%) (Fig. 2E). Multiple colors of microplastics were observed, with blue particles most abundant in water samples, while blue and white particles were predominant in oyster samples. Fiber was the dominant component among all samples, followed by foam and film respectively in water and oyster samples. Microplastic particles with sizes of 1–5 mm were more frequently observed in water samples (54.4%) than oyster samples (28.6%) ($p < 0.05$).

3.2. Microplastics in water

There were 1.47–7.61 particles/L of microplastics in the water in Maowei Sea from January to December 2018 (Fig. 3). The annual average abundance of microplastics in the inner bay (S2, 3.78 ± 0.43 particles/L) and mouth bay (S3, 3.74 ± 0.49) were slightly higher than the annual average abundance in the open bay (S4, 3.45 ± 0.39) and estuarine (S1, 3.53 ± 0.46) sites ($p > 0.05$) (Fig. S1). In addition, there was significant variation in the temporal distribution of microplastics across sampling sites ($p < 0.05$) (Fig. 3). In the estuarine nursery site (S1), the abundance of microplastics exhibited a downward trend in the first half of 2018, and then increased gradually in the second half of 2018, excluding in July and

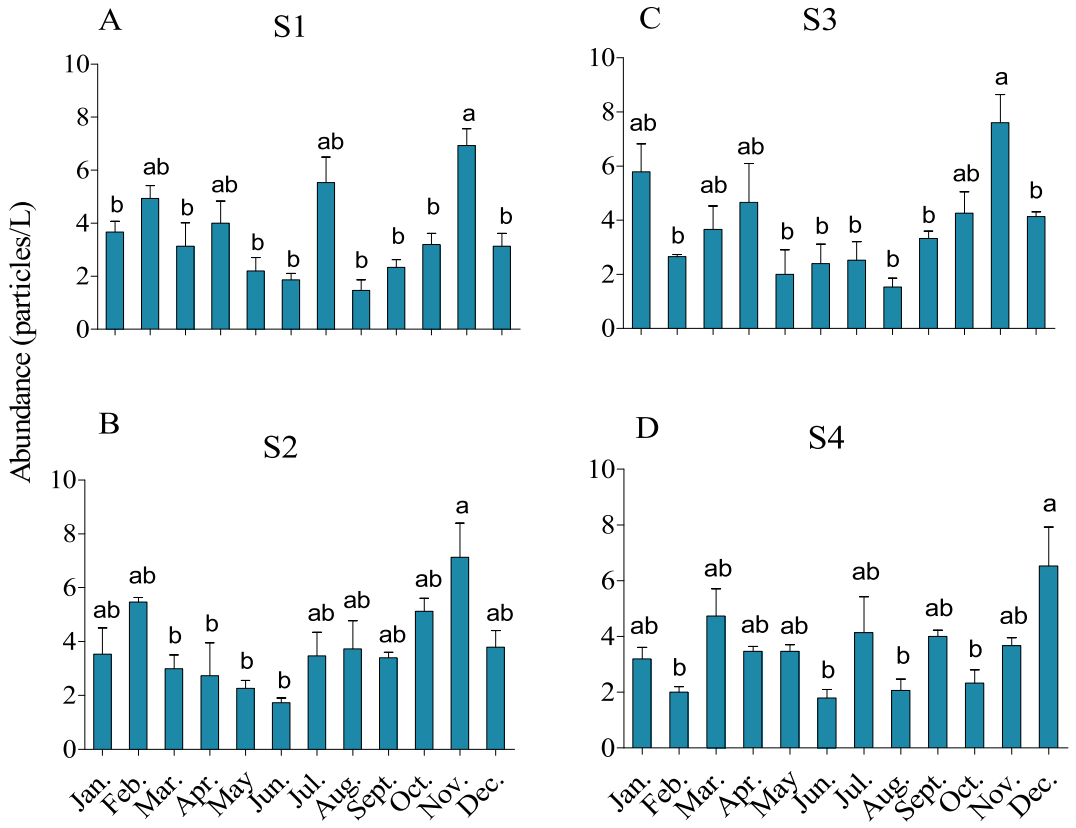

Fig. 3. Annual microplastic abundance trends in water in the estuarine region (A, S1), inner bay (B, S2), mouth bay (C, S3), and open bay (D, S4) sites in Maowei Sea. Each value represents mean ± SEM of three replicates (n = 3). Statistical analysis was conducted with repeated measures ANOVA followed by Dunnett test ($p < 0.05$). T-test was used for each two group comparisons. The letters above the scatter indicate significant differences ($p < 0.05$). If two arbitrary groups have the same letter, it means that they have no significant difference.

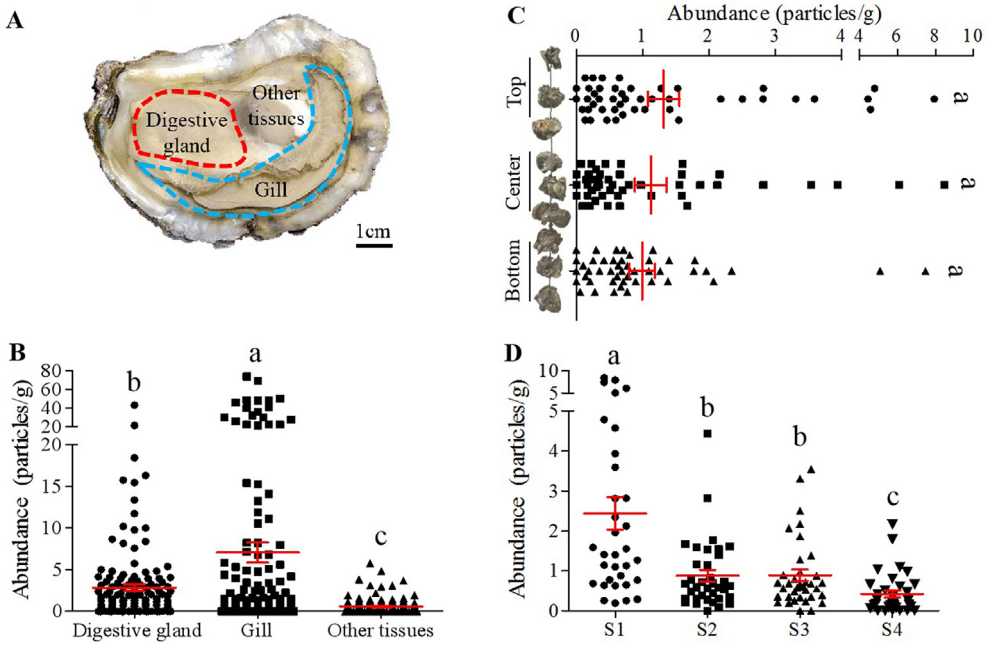

Fig. 4. Microplastic abundance in particles per wet gram in different oyster organs and the spatial distribution in Maowei Sea. (A) Oyster organ-specific microplastic analysis. (B) Comparison of microplastic abundance among different organs (n = 387). (C-D) Comparison of microplastic abundance among from different depths (C, n = 129) and sampling sites (D, n = 90–99). Each value represents mean ± SEM. Statistical analysis was conducted with repeated measures ANOVA followed by Dunnett test ($p < 0.05$). T-test was used for each two group comparisons. The letters above the scatter indicate significant differences ($p < 0.05$). If two arbitrary groups have the same letter, it means that they have no significant difference.

November. In addition, similar U-shaped time trends were observed at S2 and S3. No significant time trends were observed at S4. The abundance of microplastics peaked in Nov. at S1–S3 and increased suddenly in July at S1.

3.3. Microplastics in oyster

Microplastics were detected in the soft tissues of all oyster samples at S1, 99% samples at S2, 98% samples at S3, and 80% samples at S4. Furthermore, microplastics were observed in the digestive gland (64%), gill (55%), and other tissues (58%) of oysters (Fig. 4A) collected from Maowei Sea. More microplastics were observed in the gills (7.05 ± 1.21 particles/g), followed by digestive glands (2.84 ± 0.44 particles/g), and other tissues (0.59 ± 0.08 particles/g) based on particles per gram wet weight (Fig. 4B). However, no significant differences were observed between different oyster organs, based on particles per individual unit (Fig. S2A). No significant differences were observed in microplastic abundance among the top, center, and bottom groups both in particles per gram unit (Fig. 4C) and in particles per individual unit (Fig. S2B).

Oysters from the open bay (S4) in the Maowei Sea were the fattest, followed by those at S2–S3 and S1 ($p < 0.05$) (Fig. S2C). Conversely, microplastic pollution was most severe in oysters from S1 (2.44 ± 0.41 particles/g), followed by S2–S3 (0.88 ± 0.14 particles/g) and S4 (0.42 ± 0.09 particles/g) ($p < 0.05$) (Fig. 4D). Similar spatial variations were also observed based on particles per individual unit (Fig. S2D).

The time trends of microplastics in 2018 indicated no accumulation in oyster tissues (Fig. 5). Oysters had lower levels of microplastics in winter than in summer at S1 (excluding June) and S4 (Fig. 5A, D). The oysters at S2 were less contaminated in the first half of 2018 than in the second half of the year (Fig. 5B). Similar time trends were observed based on particles per individual unit (Fig. S3).

3.4. Relationship of microplastics in water and oysters

No significant correlation in the abundances of microplastics was observed between water and oyster following Spearman correlation analysis (r = -0.202) (Table 1). The microplastic abundance in oyster was correlated with some environmental variables of the surrounding water, such as salinity (r = -0.516), pH (r = -0.429), nutrients (r = 0.371–0.394) and total organic carbon (r = 0.482) (Table 1).

4. Discussion

4.1. Spatiotemporal distribution of microplastics in water

The average microplastic pollution level in surface water in Maowei Sea was 3.63 ± 0.22 particles/L, which is comparable to levels detected in coastal metropolis rivers in China (1.5–6.3 particles/L, Qu et al., 2018). Rivers are commonly recognized as one of the dominant pathways via which microplastics reach oceans (Law and Thompson, 2014). For land-based pollution, the concentrations will gradually decrease in space from the estuary outward. However, according to the results in this study, the microplastic concentrations at the 4 stations from the estuary to the bay did not show a downward trend. Relatively high levels of microplastics were observed at the S2 and S3 stations in the central part of the breeding area (Fig. S1). Although there were no significant

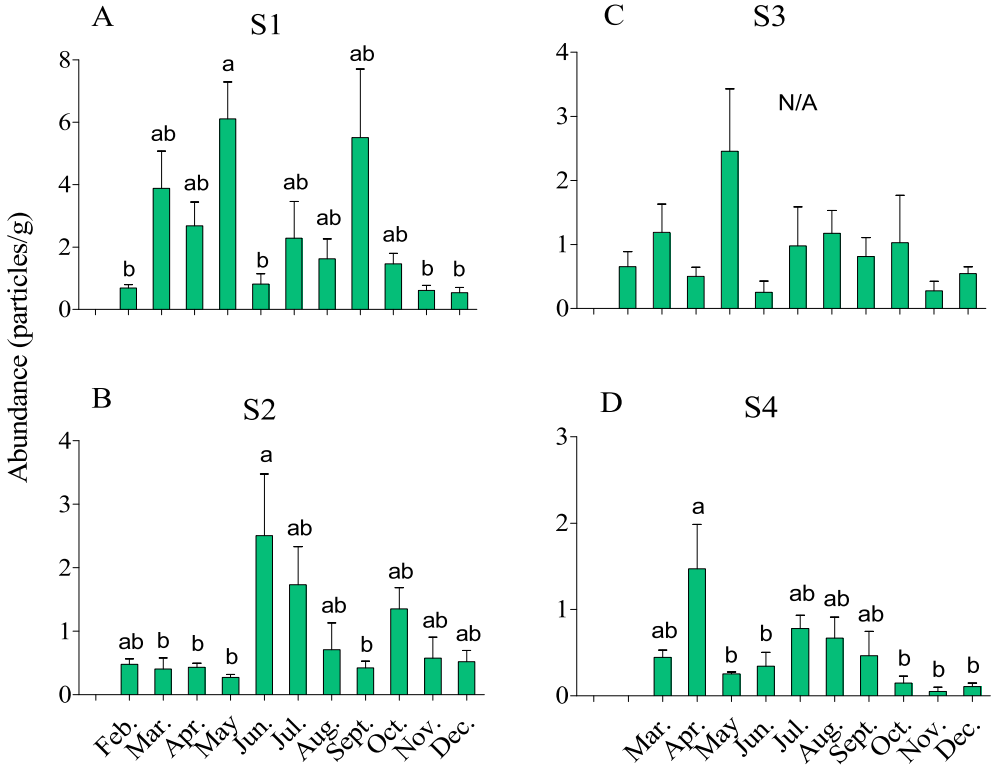

Fig. 5. Annual trends of microplastics in oysters based on particles per gram unit at the estuarine region (A, S1), inner bay (B, S2), mouth bay (C, SS), and open bay (D, S4) sites in Maowei Sea. Each value represents mean ± SEM of three replicates (n = 3). Statistical analysis was conducted with repeated measures ANOVA followed by Dunnett test ($p < 0.05$). T-test was used for each two group comparisons. The letters above the scatter indicate significant differences ($p < 0.05$). If two arbitrary groups have the same letter, it means that they have no significant difference.

Table 1
The Spearman correlation analysis of microplastic abundances and environmental variables.

	MP-W	MP-O	WT	SAL	pH	DO	Chl-a	$NH4^+$-N	TP	TN	Biomass	TOC
MP-W	1.000	−0.202	−00.377	0.301*	0.264	0.249	−0.346*	0.060	−0.087	−0.311*	−0.053	−0.265
MP-O	−0.202	1.000	0.221	−0.516**	−0.429**	−0.164	−0.012	−0.080	0.371*	0.394**	−0.042	0.482**
WT	−0.377**	0.221	1.000	−0.557**	−0.595**	−0.823**	−0.126	−0.369**	0.096	0.422**	0.462**	0.608**
SAL	0.301*	−0.516**	−0.557**	1.000	0.727**	0.441**	0.365*	0.160	−0.408**	−0.692**	−0.044	−0.772**
pH	0.264	−0.429**	−0.595**	0.727**	1.000	0.557**	0.165	0.074	−0.256	−0.552**	0.032	−0.675**
DO	0.249	−0.164	−0.823**	0.441**	0.557**	1.000	0.371**	0.283	0.018	−0.323*	−0.469**	−0.397**
Chl-a	−0.346*	−0.012	−0.126	0.365*	0.165	0.371**	1.000	0.078	−0.140	−0.066	−0.395**	−0.152
$NH4^+$-N	0.060	−0.080	−0.369**	0.160	0.074	0.283	0.078	1.000	0.122	0.066	−0.393**	−0.189
TP	−0.087	0.371*	0.096	−0.408**	−0.256	0.018	−0.140	0.122	1.000	0.472**	−0.063	0.521**
TN	−0.311*	0.394**	0.422**	−0.692**	−0.552**	−0.323*	−0.066	0.066	0.472**	1.000	−0.033	0.640**
Biomass	−0.053	−0.042	0.462**	−0.044	0.032	−0.469**	−0.395**	−0.393**	−0.063	−0.033	1.000	0.144
TOC	−0.265	0.482**	0.608**	−0.772**	−0.675**	−0.397**	−0.152	−0.189	0.521**	0.640**	0.144	1.000

Abbreviations: MP-W: microplastics in water; MP-O: microplastics in oyster; WT: water temperature; SAL: salinity; DO: dissolved oxygen; Chl-a: chlorophyll *a*; TP: total phosphorus; TN: total nitrogen; TOC: total organic carbon. * means $p < 0.05$, ** means $p < 0.01$.

differences in concentrations at our sampled locations, the data suggests that the spatial distribution of microplastics in the breeding area were not associated with land-based pollution.

U-shaped time trends from January to December 2018 were observed in the water in the estuarine region, inner bay, and mouth bay sites of Maowei Sea. The time trends had an inverse relationship with the local rainfall (Fig. S4). The rainy season in Qinzhou city is from May to September, and the microplastic abundance was low during the period. The microplastic pollution in Maowei Sea is different from that of riverine inputs. It is estimated that over 74% of riverine emissions occurring between May and October (Lebreton et al., 2017). Our previous study showed that the microplastic abundance in inflowing rivers was not higher than the abundance in Maowei Sea (Zhu et al., 2019). Furthermore, the characteristics of microplastic particles in Maowei Sea are likely to be related to textile pollution and fishery activities. For example, the common particles in Maowei Sea are 1–5 mm PET or PE fibers with diameter <20 μm (Fig. 2A), which are categorized as one type representing synthetic fibers from textiles (Wang et al., 2019; Cequier et al., 2014; Mason et al., 2016; Wang et al., 2018a,b). Polystyrene foam (Fig. 2C) is mainly related to fishery activities (Zhou, 2016), which reached about 14.3% in Maowei Sea. Many microplastic studies focused on land-based sources (Jambeck et al., 2015; Lebreton et al., 2017; Mai et al., 2019; Law et al., 2020), largely overlooked the sea-based sources, such as marine fishery activities (Lusher et al., 2017). Our work highlights the need for concern over sea-based mariculture microplastic pollution.

4.2. Accumulation and organ distribution of microplastics in oysters

The time trends of microplastics in oysters exhibited no accumulation of microplastics in oyster tissues after one-year exposure in a real environment. Our results are consistent with the findings of Ward et al. (2019), where mussels (*Mytilus edulis*) exposed to nanospheres or microspheres for 14 days at concentrations of 0.1 mg L^{-1} h^{-1} showed no accumulation of the plastics in their tissues. Mussels reportedly clear microplastics with a similar efficiency to that of clearing food items (microalgae) of similar size (Fernández and Albentosa, 2019). Our results verified no accumulation of microplastics in the real environment, based on a long-term monitoring activity.

We found more microplastics in the gills, followed by in the digestive gland and other tissues of the oysters in a real environment. Not all particles captured by the gills are ingested (Santana et al., 2018; Santana, 2015) since bivalves are selective particle feeders (Ward and Shumway 2004). Su et al. (2019) reported more microplastics in the gut than in the gills of commercial fish. In most cases, the intestinal tract would be the dominant pathway for microplastic uptake for fish. However, the dominant pathway seemed to be gills in the case of bivalves, based on our results. A reasonable explanation for the difference is the mouth feeding and filter feeding modes for fish and oysters, respectively.

4.3. Possible factors for the accumulation of microplastics by oysters

Microplastic pollution in oysters from an estuarine site was more severe than in other sites. However, no significant differences were observed among the water samples collected from the sites. Therefore, the differences in microplastics in oysters from different sites were potentially unrelated to the microplastics contaminating in the surrounding water (r = -0.202). On the contrary, some spot field studies have reported high positive correlations between mussels (Qu et al., 2018), oysters (Li et al., 2018a), and clams (Su et al., 2016), with their surrounding waters. A probable explanation is that although no accumulation was observed on the long-term scale, bivalves could ingest and accumulate microplastics over short-time scales, such as within hours (Von Moos et al., 2012; Li et al., 2019).

The microplastic abundance in oyster was correlated with some environmental variables (i.e. salinity, pH, nutrients and total organic carbon) of the surrounding water, following Spearman correlation analysis, in the present study. The microplastic levels in oysters could probably be influenced by the environmental variables. The underlying mechanisms of the accumulation of microplastic levels in oysters are complex, and the growth environment could be key factors, and not simply related to the microplastic pollution levels in the surrounding waters. Fernández and Albentosa (2019) reported that mussels cleared microplastics at the same extent than microalgae. Microplastics can not provide any nutrition to organisms and may change the microbial environment inside their bodies (Rist et al., 2017), the similar uptake model of oysters to microplastics and microalgae may pose potential damage to oyster growth and aquaculture production.

5. Conclusions

Our study is one of the first to assess the long-term trends of microplastic amounts in a mariculture zone and analyzed the relationship between surrounding water levels and accumulation of microplastics by oysters. U-shaped time trends were observed in water in Maowei Sea, the time trends had an inverse relationship with the local rainfall. The characteristics of microplastic particles

in Maowei Sea are likely to be related to textile pollution and fishery activities. We call for increased vigilance over sea-based mariculture microplastic pollution. No accumulation of microplastics in oyster tissues was observed after one-year exposure in the actual environment. Moreover, the microplastic abundance in oyster was correlated with some environmental variables (i.e. salinity, pH, nutrients and total organic carbon) of the surrounding water.

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.

Acknowledgements

This study was supported by grants from the research fund of the Natural Science Foundation of Guangxi Province (grant no. 2018JJB150065), Science and Technology Base and Talent Project of Guangxi Province (2019AC20125), Science and Technology Major Project of Guangxi Province (AA17129001; AA17204095-10), Fundamental Research Funds for the Central Universities (2019B05514), and the Open Research Fund Program of Guangxi Key Lab of Mangrove Conservation and Utilization (GKLMC-201901).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2021.116450.

References

Auta, H.S., Emenike, C.U., Fauziah, S.H., 2017. Distribution and importance of microplastics in the marine environment: a review of the sources, fate, effects, and potential solutions. Environ. Int. 102, 165–176.
Bevington, P.R., Robinson, D.K., Blair, J.M., Mallinckrodt, A.J., McKay, S., 1993. Data reduction and error analysis for the physical sciences. Comput. Phys. 7 (4), 415–416.
Browne, M.A., Crump, P., Niven, S.J., Teuten, E., Tonkin, A., Galloway, T., Thompson, R., 2011. Accumulation of microplastic on shorelines worldwide: sources and sinks. Environ. Sci. Technol. 45 (21), 9175–9179.
Castro, R.O., Silva, M.L., Marques, M.R.C., de Araújo, F.V., 2016. Evaluation of microplastics in Jurujuba Cove, Niterói, RJ, Brazil, an area of mussels farming. Mar. Pollut. Bull. 110 (1), 555–558.
Cequier, E., Ionas, A.C., Covaci, A., Marcé, R.M., Becher, G., Thomsen, C., 2014. Occurrence of a broad range of legacy and emerging flame retardants in indoor environments in Norway. Environ. Sci. Technol. 48 (12), 6827–6835.
Chen, M., Jin, M., Tao, P., Wang, Z., Xie, W., Yu, X., Wang, K., 2018. Assessment of microplastics derived from mariculture in Xiangshan Bay, China. Environ. Pollut. 242, 1146–1156.
Chen, Q., Zhang, H., Allgeier, A., Zhou, Q., Ouellet, J.D., Crawford, S.E., Hollert, H., 2019. Marine microplastics bound dioxin-like chemicals: model explanation and risk assessment. J. Hazard Mater. 364, 82–90.
Cheung, P.K., Fok, L., Hung, P.L., Cheung, L.T., 2018. Spatio-temporal comparison of neustonic microplastic density in Hong Kong waters under the influence of the Pearl River Estuary. Sci. Total Environ. 628, 731–739.
Cox, K.D., Covernton, G.A., Davies, H.L., Dower, J.F., Juanes, F., Dudas, S.E., 2019. Human consumption of microplastics. Environ. Sci. Technol. 53 (12), 7068–7074.
Dris, R., Gasperi, J., Rocher, V., Saad, M., Renault, N., Tassin, B., 2015. Microplastic contamination in an urban area: a case study in Greater Paris. Environ. Chem. 12 (5), 592–599.
Eo, S., Hong, S.H., Song, Y.K., Han, G.M., Shim, W.J., 2019. Spatiotemporal distribution and annual load of microplastics in the Nakdong River, South Korea. Water Res. 160, 228–237.
Feng, Z., Zhang, T., Li, Y., He, X., Wang, R., Xu, J., Gao, G., 2019. The accumulation of microplastics in fish from an important fish farm and mariculture area, Haizhou Bay, China. Sci. Total Environ. 696, 133948.
Fernández, B., Albentosa, M., 2019. Insights into the uptake, elimination and accumulation of microplastics in mussel. Environ. Pollut. 249, 321–329.
Fok, L., Cheung, P.K., 2015. Hong Kong at the Pearl River Estuary: a hotspot of microplastic pollution. Mar. Pollut. Bull. 99 (1–2), 112–118.
Frere, L., Paul-Pont, I., Rinnert, E., Petton, S., Jaffré, J., Bihannic, I., Huvet, A., 2017. Influence of environmental and anthropogenic factors on the composition, concentration and spatial distribution of microplastics: a case study of the Bay of Brest (Brittany, France). Environ. Pollut. 225, 211–222.
Frias, J.P.G.L., Nash, R., 2019. Microplastics: finding a consensus on the definition. Mar. Pollut. Bull. 138, 145–147.
Gago, J., Filgueiras, A., Pedrotti, M.L., Caetano, M., Frias, J., 2018. Standardised protocol for monitoring microplastics in seawater. JPI-Oceans BASEMANproject.
GESAMP, 2015. Sources, fate and effects of microplastics in the marine environment: part 2 of a global assessment. Kershaw, P.J., Rochman, C.M. Rep. Stud. GESAMP No. 90(96).
Gosling, E., 2008. Bivalve Molluscs: Biology, Ecology and Culture. John Wiley & Sons.
Hollman, P.C., Bouwmeester, H., Peters, R.J.B., 2013. Microplastics in aquatic food chain: sources, measurement, occurrence and potential health risks (No. 2013.003). Rikilt-Institute of Food Safety.
Hu, L., Chernick, M., Hinton, D.E., Shi, H., 2018. Microplastics in small waterbodies and tadpoles from Yangtze River Delta, China. Environ. Sci. Technol. 52 (15), 8885–8893.
Hurley, R.R., Woodward, J.C., Rothwell, J.J., 2017. Ingestion of microplastics by freshwater tubifex worms. Environ. Sci. Technol. 51 (21), 12844–12851.
Jambeck, J.R., Geyer, R., Wilcox, C., Siegler, T.R., Perryman, M., Andrady, A., Law, K.L., 2015. Plastic waste inputs from land into the ocean. Science 347 (6223), 768–771.
Koelmans, A.A., Besseling, E., Foekema, E., Kooi, M., Mintenig, S., Ossendorp, B.C., Scheffer, M., 2017. Risks of plastic debris: unravelling fact, opinion, perception, and belief. Environ. Sci. Technol. 51 (20), 11513–11519.
Lambert, S., Scherer, C., Wagner, M., 2017. Ecotoxicity testing of microplastics: considering the heterogeneity of physicochemical properties. Integrated Environ. Assess. Manag. 13 (3), 470–475.
Law, K.L., Thompson, R.C., 2014. Microplastics in the seas. Science 345 (6193), 144–145.
Law, K.L., Starr, N., Siegler, T.R., Jambeck, J.R., Mallos, N.J., Leonard, G.H., 2020. The United States' contribution of plastic waste to land and ocean. Science advances, 6(44), eabd0288.
Lebreton, L.C., Van Der Zwet, J., Damsteeg, J.W., Slat, B., Andrady, A., Reisser, J., 2017. River plastic emissions to the world's oceans. Nat. Commun. 8, 15611.
Li, H.X., Ma, L.S., Lin, L., Ni, Z.X., Xu, X.R., Shi, H.H., Rittschof, D., 2018a. Microplastics in oysters saccostrea cucullata along the pearl river estuary, China. Environ. Pollut. 236, 619–625.
Li, J., Green, C., Reynolds, A., Shi, H., Rotchell, J.M., 2018b. Microplastics in mussels sampled from coastal waters and supermarkets in the United Kingdom. Environ. Pollut. 241, 35–44.
Li, J., Lusher, A.L., Rotchell, J.M., Deudero, S., Turra, A., Bråte, I.L.N., Shi, H., 2019. Using mussel as a global bioindicator of coastal microplastic pollution. Environ. Pollut. 244, 522–533.
Lima, A.R.A., Costa, M.F., Barletta, M., 2014. Distribution patterns of microplastics within the plankton of a tropical estuary. Environ. Res. 132, 146–155.
Lusher, A., Hollman, P., Mendoza-Hill, J., 2017. Microplastics in fisheries and aquaculture: status of knowledge on their occurrence and implications for aquatic organisms and food safety. FAO Fisher. Aquacult. Techn. Pap. (615).
Macfadyen, G., Huntington, T., Cappell, R., 2009. Abandoned, lost or otherwise discarded fishing gear. In: UNEP Regional Seas Reports and Studies, No. 185; FAO Fisheries and Aquaculture Technical Paper. UNEP/FAO, Rome, p. 115.
Mai, L., You, S.N., He, H., Bao, L.J., Liu, L.Y., Zeng, E.Y., 2019. Riverine microplastic pollution in the Pearl River Delta, China: are modeled estimates accurate? Environ. Sci. Technol. 53 (20), 11810–11817.
Mason, S.A., Garneau, D., Sutton, R., Chu, Y., Ehmann, K., Barnes, J., Rogers, D.L., 2016. Microplastic pollution is widely detected in US municipal wastewater treatment plant effluent. Environ. Pollut. 218, 1045–1054.
McLusky, D.S., Elliott, M., 2004. The estuarine ecosystem: ecology, threats and management. OUP Oxford.
Qu, X., Su, L., Li, H., Liang, M., Shi, H., 2018. Assessing the relationship between the abundance and properties of microplastics in water and in mussels. Sci. Total Environ. 621, 679–686.
Rist, S., Baun, A., Hartmann, N.B., 2017. Ingestion of micro-and nanoplastics in Daphnia magna–Quantification of body burdens and assessment of feeding rates and reproduction. Environ. Pollut. 228, 398–407.
Rist, S., Steensgaard, I.M., Guven, O., Nielsen, T.G., Jensen, L.H., Møller, L.F., Hartmann, N.B., 2019. The fate of microplastics during uptake and depuration phases in a blue mussel exposure system. Environ. Toxicol. Chem. 38 (1), 99–105.
Rocha-Santos, T., Duarte, A.C., 2015. A critical overview of the analytical approaches to the occurrence, the fate and the behavior of microplastics in the environment. Trac. Trends Anal. Chem. 65, 47–53.
Rochman, C.M., Browne, M.A., Underwood, A.J., Franeker, Van, Thompson, R.C., Amaral-Zettler, L.A., 2016. The ecological impacts of marine debris: unraveling the demonstrated evidence from what is perceived. Ecology 97 (2), 302–312.
Rodrigues, M.O., Abrantes, N., Gonçalves, F.J.M., Nogueira, H., Marques, J.C., Gonçalves, A.M.M., 2018. Spatial and temporal distribution of microplastics in water and sediments of a freshwater system (Antuã River, Portugal). Sci. Total Environ. 633, 1549–1559.
Santana, M.F.M., 2015. Effects of microplastics contamination on marine biota (Doctoral dissertation, Universidade de São Paulo).
Santana, M.F., Moreira, F.T., Pereira, C.D., Abessa, D.M., Turra, A., 2018. Continuous exposure to microplastics does not cause physiological effects in the cultivated

mussel Perna perna. Arch. Environ. Contam. Toxicol. 74 (4), 594–604.
Setälä, O., Fleming-Lehtinen, V., Lehtiniemi, M., 2014. Ingestion and transfer of microplastics in the planktonic food web. Environ. Pollut. 185, 77–83.
Shim, W.J., Hong, S.H., Eo, S., 2018. Marine microplastics: abundance, distribution, and composition. In: Microplastic Contamination in Aquatic Environments. Elsevier, pp. 1–26.
Su, L., Deng, H., Li, B., Chen, Q., Pettigrove, V., Wu, C., Shi, H., 2019. The occurrence of microplastic in specific organs in commercially caught fishes from coast and estuary area of east China. J. Hazard Mater. 365, 716–724.
Su, L., Xue, Y., Li, L., Yang, D., Kolandhasamy, P., Li, D., Shi, H., 2016. Microplastics in taihu lake, China. Environ. Pollut. 216, 711–719.
Van Cauwenberghe, L., Claessens, M., Vandegehuchte, M.B., Janssen, C.R., 2015. Microplastics are taken up by mussels (Mytilus edulis) and lugworms (Arenicola marina) living in natural habitats. Environ. Pollut. 199, 10–17.
Von Moos, N., Burkhardt-Holm, P., Köhler, A., 2012. Uptake and effects of microplastics on cells and tissue of the blue mussel Mytilus edulis L. after an experimental exposure. Environ. Sci. Technol. 46 (20), 11327–11335.
Wagner, M., Scherer, C., Alvarez-Muñoz, D., Brennholt, N., Bourrain, X., Buchinger, S., Rodriguez-Mozaz, S., 2014. Microplastics in freshwater ecosystems: what we know and what we need to know. Environ. Sci. Eur. 26 (1), 1–9.
Wang, T., Zou, X., Li, B., Yao, Y., Li, J., Hui, H., Wang, C., 2018a. Microplastics in a wind farm area: a case study at the Rudong Offshore wind farm, yellow sea, China. Mar. Pollut. Bull. 128, 466–474.
Wang, T., Zou, X., Li, B., Yao, Y., Zang, Z., Li, Y., Wang, W., 2019. Preliminary study of the source apportionment and diversity of microplastics: taking floating microplastics in the South China Sea as an example. Environ. Pollut. 245, 965–974.
Wang, W.X., 2016. Bioaccumulation and biomonitoring. In: Marine Ecotoxicology. Academic Press, pp. 99–119.
Wang, W., Yuan, W., Chen, Y., Wang, J., 2018b. Microplastics in surface waters of dongting lake and hong lake, China. Sci. Total Environ. 633, 539–545.
Ward, J.E., Shumway, S.E., 2004. Separating the grain from the chaff: particle selection in suspension-and deposit-feeding bivalves. J. Exp. Mar. Biol. Ecol. 300 (1–2), 83–130.
Ward, J.E., Rosa, M., Shumway, S.E., 2019. Capture, ingestion, and egestion of microplastics by suspension-feeding bivalves: a 40-year history. Anthropocene Coasts 2 (1), 39–49.
Xu, X.Y., Lee, W.T., Chan, A.K.Y., Lo, H.S., Shin, P.K.S., Cheung, S.G., 2017. Microplastic ingestion reduces energy intake in the clam Atactodea striata. Mar. Pollut. Bull. 124 (2), 798–802.
Yang, D., Shi, H., Li, L., Li, J., Jabeen, K., Kolandhasamy, P., 2015. Microplastic pollution in table salts from China. Environ. Sci. Technol. 49 (22), 13622–13627.
Zhang, H., 2017. Transport of microplastics in coastal seas. Estuarine. Coast. Shelf Sci. 199, 74–86.
Zhang, Q., Zhao, Y., Du, F., Cai, H., Wang, G., Shi, H., 2020. Microplastic fallout in different indoor environments. Environ. Sci.Technol.
Zhou, Q., 2016. Occurrences and ecological risks of microplastics in the typical coastal Beaches and seas. master thesis Yantai Inst. Coast. Zone Res., Chin. Acad. Sci. Yantai (in Chinese).
Zhu, J., Zhang, Q., Li, Y., Tan, S., Kang, Z., Yu, X., Shi, H., 2019. Microplastic pollution in the Maowei Sea, a typical mariculture bay of China. Sci. Total Environ. 658, 62–68.