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Spatio-temporal features of microplastics pollution in macroalgae growing in an important mariculture area, China

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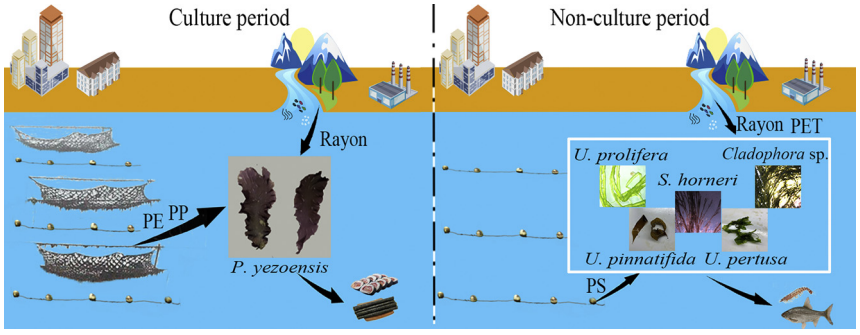
HIGHLIGHTS

- Mariculture releases a large amount of plastics into the environment annually.
- Macroalgae can accumulate microplastics via diverse mechanisms.
- Mariculture exacerbates microplastics pollution in macroalgae.
- Macroalgae can be ideal bioindicators for microplastics pollution.

GRAPHICAL ABSTRACT



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ABSTRACT

Macroalgae are being consumed by a growing number of people as functional food. Therefore, they are intensively cultivated to meet the rising demand. Mariculture is a potential source of microplastics (MPs). However, as a potential source of microplastics, little is known regarding the MPs pollution in macroalgae of open sea macriculture. Here we investigated the MPs characteristics in macroalgae in three sections of Haizhou Bay, an important mariculture area in China, during *Pyropia* culture (*Pyropia yezoensis*) and non-culture periods (*Ulva prolifera*, *Sargassum horneri*, *Cladophora* sp., *Undaria pinnatifida*, *Ulva pertusa*). It was found that *P. yezoensis* during the culture period had higher MPs abundance (0.17 ± 0.08 particles g^{-1}fresh weight) than other macroalgae (0.12 ± 0.09 particles g^{-1} fresh weight) during the non-culture period, particularly for the nearshore sections. There were more fiber MPs in *P. yezoensis* (90.43%) in culture period compared to macroalgae (84.46%) in non-culture period. Highly similar spectrum of plastics in culture gears and macroalgae was verified. *Pyropia* culture gears released about 1,037 tons plastics into the environment annually and the MPs abundances in seawater during the culture and non-culture periods were 1.04 ± 0.32 and 1.86 ± 0.49 particles L^{-1}, respectively. The gap of MPs abundance between the two periods can be attributed to the tremendous trapping by massive biomass of *P. yezoensis* during the culture period and the continuous plastic release during the non-culture period. This

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study indicates that culture gears of macroalgae could be an important MPs source and the MPs can be transferred to human by edible macroalgae, and meanwhile macroalgae may be ideal biomonitors for MPs pollution in seawater due to their unbiased trapping and immovability.

1. Introduction

Plastics have been used more and more in industrial production and daily life since it had been produced (Barnes et al., 2009; Haward, 2018), and the annual global plastics production has exceeded 359 million tons in 2018 (Plastics Europe, 2019). A large amount of plastic garbage enters the ocean due to improper handling by humans. As a result, plastic materials account for 80–85% of total marine waste (Auta et al., 2017). It had been reported that severe plastic pollution occurred in Pacific Ocean (Hipfner et al., 2018), Amazon River estuary (Pegado et al., 2018), Mediterranean Sea (Bellas et al., 2016; Romeo et al., 2016), and the Arctic Ocean (Morgana et al., 2018). Nowadays, plastic pollution is posing significant impacts on the marine ecosystem (Lusher et al., 2015; Zalasiewicz et al., 2016; Galloway et al., 2017). Microplastics (MPs) are described as any pieces of plastics in size <5 mm and even smaller particles in the environment (Thompson et al., 2004). Compared to large plastics, MPs are easier to be dispersed throughout the oceans due to its smaller particle size. Consequently, MPs are ubiquitous in the oceans and available for marine organisms in different depths and accidental ingestion by marine organisms (Moore, 2008; Lusher et al., 2015; Hu et al., 2018). Furthermore, MPs can adsorb and carry more toxic matters, such as heavy metal and persistent organic pollutant, thanks to its larger specific surface area (Wang et al., 2018a, 2019a). MPs have been shown negative effects on physiological performance of marine organisms (Wang et al., 2018a, 2018b). Therefore, MPs have become a potentially enormous challenge to the global marine environment and ecosystem (Guzzetti et al., 2018; Haward, 2018).

China is the biggest country for plastic production and consumption and deemed to discharge most plastics into the ocean (Jambeck et al., 2015). Therefore, a number of studies for MPs pollution in Chinese coastal areas have been carried out recently (Zhao et al., 2014; Yu et al., 2016; Jabeen et al., 2017; Zhu et al., 2018; Wang et al., 2019b). It seems that MPs pollution in Chinese seas is more serious compared to other areas in the world (Zhu et al., 2018; Wang et al., 2019b). For example, Feng et al. (2019) found that the abundance of MPs in fish in the Yellow Sea is higher than those in North Pacific Central Gyre (Boerger et al., 2010), the Spanish Atlantic and Mediterranean coasts (Bellas et al., 2016), the Northeast Atlantic around Scotland (Murphy et al., 2017) and the Saudi Arabian Red Sea coast (Baalkhuyur et al., 2018). The present studies on MPs focus on their distribution in seawater, sediment and marine animals (Yokota et al., 2017; De Sá et al., 2018; Mishra et al., 2019; Zhang et al., 2019), and little is known about MPs trapping in macroalgae.

Macroalgae are the main contributors to coastal primary production and meanwhile many species have also been consumed by human as sea vegetables due to their rich content of dietary fiber, protein, vitamins and minerals (Gao et al., 2018a; Mouritsen et al., 2019). In the Far East and Pacific, there has been a long tradition of consuming macroalgae as food and macroalgae has formed part of the daily diet in China, Japan and Korea for centuries (Roohinejad et al., 2017; Gao et al., 2018b). The demand of macroalgae far outstrips supply and therefore intensive macroalgae culture is being conducted in Asian countries. Culture gears for macroalgae including net curtain, rope and buoyant ball employ a variety of plastic materials. The plastic gear is a potential source of MPs to the coastal environment, but relevant data on the impacts of macroalgae culture in open sea in terms of MPs pollution are still very scarce.

Haizhou Bay, open to the Yellow Sea, has a long-term mariculture history for *Pyropia yezoensis*, accounting for about 50% of production in China and the cultivation (58, 589 ha) has been expanded to open sea (Lu et al., 2018; Xu et al., 2019) (Fig. 1). Therefore, this study hypothesized that mariculture may generate MPs pollution to environment and macroalgae, and assessed the MPs distribution in seawater, and MPs trapping in *P. yezoensis* during the culture period and that in other macroalgae attached to culture gear during the non-culture period in Haizhou Bay.

2. Materials and methods

2.1. Sample collection and preparation

Macroalgae samples were collected in *Pyropia* culture area of Haizhou Bay in the Yellow Sea of China in February and June 2019. These two months represent *P. yezoensis* culture period and non-culture period, respectively. The sampling was conducted in nine

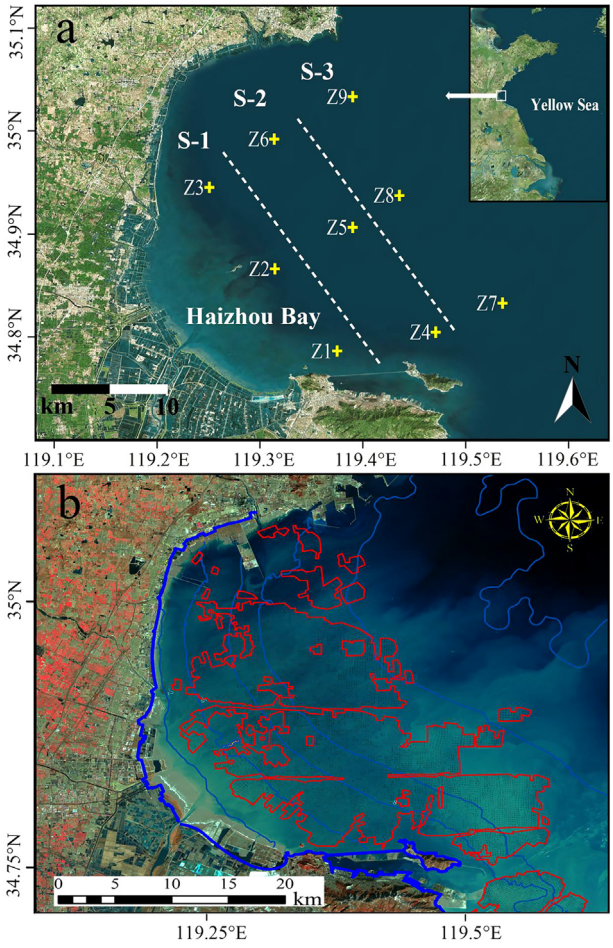

Fig. 1. Geographic position of nine sampling sites (a) and *Pyropia* culture area in Haizhou Bay (b). The sea area for *Pyropia* culture is lined out in red, being 58, 589 ha in Haizhou Bay and adjacent waters. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

stations of three sections as shown in Fig. 1 and Table S1. In February, only *P. yezoensis* was collected because other macroalgae were not available on the culture gears, while in June when *P. yezoensis* does not grow, *Ulva Prolifera*, *Sargassum horneri*, *Cladophora* sp., *Undaria pinnatifida* and *Ulva pertusa* on the ropes of *Pyropia* culture were collected. Three parallel samples for each species at each station were collected and biomass for each sample was approximately 100 g (Fig. 1). Following sampling, macroalgae samples were quickly sealed with aluminum foil bags and transferred in a cooler (−5 °C) to the laboratory where they were stored at −20 °C pending processing and analysis. About 30 L of surface seawater at each station was sampled at a 30 cm water depth by Niskin water sampler and poured through a 33 μm steel sieve to measure MPs abundance in seawater. The particles on the steel sieve were washed into a clean glass bottle with filtered deionized water and stored with 5% formalin solution pending analysis (Zhao et al., 2014, 2015). Salinity and pH of seawater at sampling sites were also measured using portable salinity meter (Thermo scientific, Eutech Salt 6+) and pH meter (Oakton, pHTestr 20).

2.2. Quality control and assurance

In order to minimize the plastics contamination from sampling tools, the metal scoop net was used to collect macroalgae samples. After sampling, macroalgae samples were quickly placed in aluminum foil bags to reduce air pollution and transported to the laboratory as soon as possible. Appropriate precautions were taken in the laboratory to avoid or reduce plastic contamination, such as minimizing the number of operators and reducing air circulation. The hands and forearms of operators were scrubbed three times before experiment and operators were required to wear white cotton lab coats, disposable nitrile gloves and masks throughout the sample handling process. Before being used, all instruments were thoroughly washed with 75% alcohol and all chemicals including deionized water were filtered through 2.7 μm glass microfiber filters (Whatman, grade GF/D). Preparation of solution, digestion of macroalgae and observation of filter membrane were always carried out in a laminar flow cabinet (SW-CJ-2F, Suzhou, China). Five blanks without macroalgae samples were prepared during each batch of sample processing to determine the degree of MPs contamination under laboratory conditions.

2.3. Sample digestion

Seawater on the surface of macroalgae was removed by absorbent tissues and the fresh weight of macroalgae was determined using a precision electronic balance (BS124S, Sartorius, Beijing). Then macroalgae samples (80–120 g) were immediately placed into a 1000 mL clean beaker and digested by 100–120 mL Fenton reagent containing 30% (*v*/v) H_2O_2 and catalyst solution (20 g of $FeSO_4 \cdot 7H_2O$ in 1 L of filtered RO water) in oscillation incubators (DKZ-3, Shanghai Yiheng, China) at 40 °C with 60 rpm for 24–48 h until they were completely dissolved (Fenton, 1894; Tagg et al., 2017; Hurley et al., 2018). The digestion solution was transferred and filtered through a 2.7 μm glass microfiber filter (Whatman, grade GF/D) using a filtration unit with one Büchner funnel (AP-01P, Autoscience, China). Then, the filter membrane was placed in a clean Petri dish with lid and dried at room temperature for further analysis.

2.4. Identification of plastics

The suspected plastic particles were isolated, photographed and measured by utilizing Nikon SMZ 1500 N (Japan) stereo microscope with charge-coupled device (CCD) camera at the largest cross section. The selected MPs were categorized according to their shape, size and colour. Approximately 25% of the total membranes with filtered substances in each site were randomly selected (corresponding to 282 verified MPs particles) and identified by a micro-Fourier Transformed Infrared Spectroscope (μ-FT-IR) (Nicolet iN10, Thermo Fisher Scientific, USA). Materials of culture gears were also taken for polymer identification. The transmittance mode was used to measure MPs. The spectrum range was set from 650 to 4000 cm^{-1} with 64 co-scans at a resolution of 8 cm^{-1}, and the aperture ranged from 50 × 50 mm to 150 × 150 mm depended on the size of particles. The OMNIC software was used for the identification of polymer by comparing the samples to a standard spectral library. Polymers showing a match degree of >70% with reference to standard spectra were validated (Thompson et al., 2004; Cai et al., 2019). All data used to calculate abundance were based on the number of plastics observed through visual recognition.

2.5. Observation of the approaches of plastics trapping in/on macroalgae

About 80–120 g macroalgae samples were randomly selected from each sampling site and observed directly using Nikon SMZ 1500 N (Japan) stereo microscope with charge-coupled device (CCD) camera to study the trapping approaches of MPs in macroalgae. All observed and isolated potential MPs were also identified with a μ-FT-IR (Nicolet iN10, Thermo Fisher Scientific, USA).

2.6. Assessment of plastics released from gears for macroalgae raft cultivation

To estimate the release amount of plastics from culture gears, relevant data were collected through field investigation and literature survey. The amount of released plastics from culture gears was calculated by the following formula: $P = U \times (A/K) \times R$, where P represents released plastics (t/year); U represents the usage amount of plastic gears per unit area of net curtain for nori culture (kg/ha), which was obtained based on field investigation; A represents the total sea area for cultivation (estimated from remote sensing data, Xu et al., 2019), K represents the coefficient showing the relationship between total sea area for cultivation and the area of net curtain for nori culture, with 1.6 being

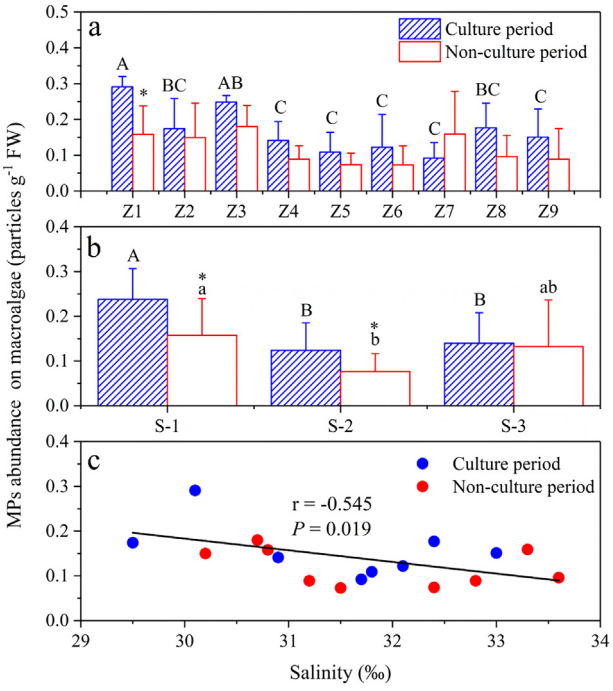

Fig. 2. Abundance of microplastics (mean ± SD) on macroalgae from the nine sites (a) and three sections (b) and the correlation between MPs abundance on macroalgae and seawater salinity (c) during the culture and non-culture periods of *P. yezoensis*. Different capital and lowercase letters above the bars indicate significant differences among sites (a) or sections (b) during the culture and non-culture periods respectively. Asterisks represent significant differences between the culture and non-culture periods ($P < .05$).

Table 1
Chemical parameters of seawater at nine sampling stations in two different periods. February (Feb) and June (Jun) represent culture and non-culture periods, respectively.

Sites	Z1	Z2	Z3	Z4	Z5	Z6	Z7	Z8	Z9
pH (Feb)	8.28	8.31	8.30	8.34	8.41	8.32	8.29	8.33	8.35
pH (Jun)	8.22	8.23	8.28	8.26	8.32	8.35	8.32	8.39	8.34
Salinity (Feb, ‰)	30.1	29.5	29.8	30.9	31.8	32.1	31.7	32.4	33.0
Salinity (Jun, ‰)	30.8	30.2	30.7	31.2	32.4	31.5	33.3	33.6	32.8

used in this study according to field investigation; and R represents the loss rate of plastic gear per year (%/year) that was determined by the mass change of plastic gear during 5-year usage.

2.7. Data analysis

The data were analyzed using the software SPSS v.23. The data under every treatment conformed to a normal distribution (Shapiro-Wilk, $P > .05$) and the variances could be considered equal (Levene's test, $P > .05$). Two-way analysis of variance (ANOVA) was conducted to assess the effect of location and culture period on the abundance of MPs in macroalgae. Curve fitting was conducted to analyze the relationship between size distribution and number of MPs. One-way ANOVA was conducted to assess differences of MPs abundance among macroalgae or seawaters. Least significant difference was conducted for ANOVA post hoc analysis. A confidence interval of 95% was set for all tests.

3. Results

3.1. Spatio-temporal distribution and composition analysis of MPs in macroalgae

The MPs abundance on macroalgae in different areas of Haizhou Bay was first shown (Fig. 2). Two-way ANOVA shows that both station ($F_{(8, 57)} = 2.860$, $P = .010$) and season ($F_{(1, 57)} = 6.284$, $P = .015$) affected MPs abundance but they did not have an interactive effect ($F_{(8, 57)} = 1.029$, $P = .425$). During the culture period (Fig. 2a), the highest MPs abundance was found at station Z1 (0.29 ± 0.03 particles g^{-1} FW), followed by station Z3 (0.25 ± 0.02 particles g^{-1} FW). During the non-culture period, the differences among different stations were statistically insignificant (One-way ANOVA, $F_{(8, 47)} = 1.331$, $P = .257$). The MPs abundance during the culture period was higher than that during the non-culture period for most stations although the difference was statistically significant only at station Z1. Section ($F_{(2, 69)} = 9.983$, $P < .001$) and season ($F_{(1, 69)} = 6.075$, $P = .016$) also affected MPs abundance (Fig. 2b). During the culture period, MPs abundance at S-1 (0.24 ± 0.07 particles g^{-1} FW) was significantly higher than those at S-2 (0.12 ± 0.06 particles g^{-1} FW) and S-3 (0.14 ± 0.07 particles g^{-1} FW). During the non-culture period, the lowest MPs abundance occurred at S-2 (0.08 ± 0.04 particles g^{-1} FW), with insignificant difference between S-1 (0.16 ± 0.08 particles g^{-1} FW) and S-3 (0.13 ± 0.10 particles g^{-1} FW). MPs abundance during the culture period was higher than that during the non-culture period for each section although the difference at S-3 was not statistically significant. The chemical parameters (pH and salinity) of seawater in the sampling areas were also investigated and pH and salinity ranged 8.22–8.41 and 29.8–33.6‰, respectively (Table 1). Salinity of seawater at S-1 was lower than that at S-2 and S-3. Furthermore, the correlations between MPs abundance on macroalgae/in seawater and seawater pH/salinity were analyzed (Table S2) and a negative correlation between MPs abundance on macroalgae and salinity was observed ($r = -0.545$, $P = .019$, Fig. 2c).

The plastics release from culture gears in Haizhou Bay was also investigated (Table 2). The culture gears include rope, net curtain, big floating bowl and floating ball, all of which are made of plastic. Rope (25, 559 t) contributes the most plastic materials, followed by net curtain (14, 830 t) and big floating bowl (5, 090 t). Big floating bowl had the highest loss rate (4.9%/year) while net curtain had the lowest value (1.2%/year). The main release of plastics was from ropes (14.66 kg/ha/year) and big floating bowl (6.81 kg/ha/year). Due to the degradation of culture gears, the annual plastics release reached 1, 037 t in Haizhou Bay.

To verify the effect of *Pyropia* culture on MPs pollution in macroalgae, plastics composition in culture gear and macroalgae was analyzed. Net curtain is made of PE, rope is made of PE and PP while floating ball is made of PS (Fig. 3). Highly similar spectra were also found in MPs trapped by macroalgae. The main polymer absorption band in culture gears and macroalgae showed the same except for the wavenumbers between 1010 and 1050 cm^{-1} where MPs in macroalgae had deeper peaks. In addition, the shape and colour in plastics between culture gear and macroalgae were quite similar as well. These results indicate that plastics released from culture gear can be trapped by macroalgae directly.

3.2. Characteristics of MPs in macroalgae during the culture and non-culture periods

As shown in Fig. 4, MPs with diverse shapes, colours and materials were found in macroalgae. In terms of shape of MPs in macroalgae (Fig. 4a, b), during the culture period, over 90% MPs were fiber, and the rest minor part was mainly constituted by foam (7.23%) and film (1.70%). During the non-culture period, the dominant form was still fiber although the percentage decreased to 84.46%; the percentage of foam increased to 12.23% and film only contributed 0.66%. The dominant colours were blue (24.68%) and transparent (23.40%) during the culture period while they were white (31.07%) and transparent (21.65%) during the non-culture period (Fig. 4c, d). Eleven polymer types (PE, Rayon, PP, PS, PET, PE-PP, CP, Nylon, PAN, PS&PAN&PMMA, and PMMA) in total were found on macroalgae (Fig. 4e, f) and the spectra of eight main types were shown (Fig. S1), which had matching degrees of >70% with reference to standard spectra. The most material was PE (39.31%) during the culture period (Fig. 4e), followed by Rayon (17.93%) and PP (12.41%); while PET led the MPs materials (27.01%), followed by Rayon (21.17%) and PS (14.60%) during the non-culture period (Fig. 4f).

Table 2
Plastic gears used for nori raft culture in Haizhou Bay and related plastics release.

Name of gears	Material	Usage amount (kg/ha)[a]	Total amount (t)[b]	Loss rate (%/year)[c]	Plastics release rate (kg/ha/year)[b]	Released plastics (t/year)[b]
Rope	PP or PE	698	25,559	2.1	14.66	537
Net curtain	PE	405	14,830	1.2	4.86	178
Big floating bowl	PS	139	5090	4.9	6.81	249
Floating ball	PS	9	330	2.2	1.98	73
Total	/	1251	45,809	/	28.31	1037

[a] The data were obtained by field investigation;
[b] The data were calculated according to formula;
[c] The loss rate was determined based on the mass changes of culture gear with time.

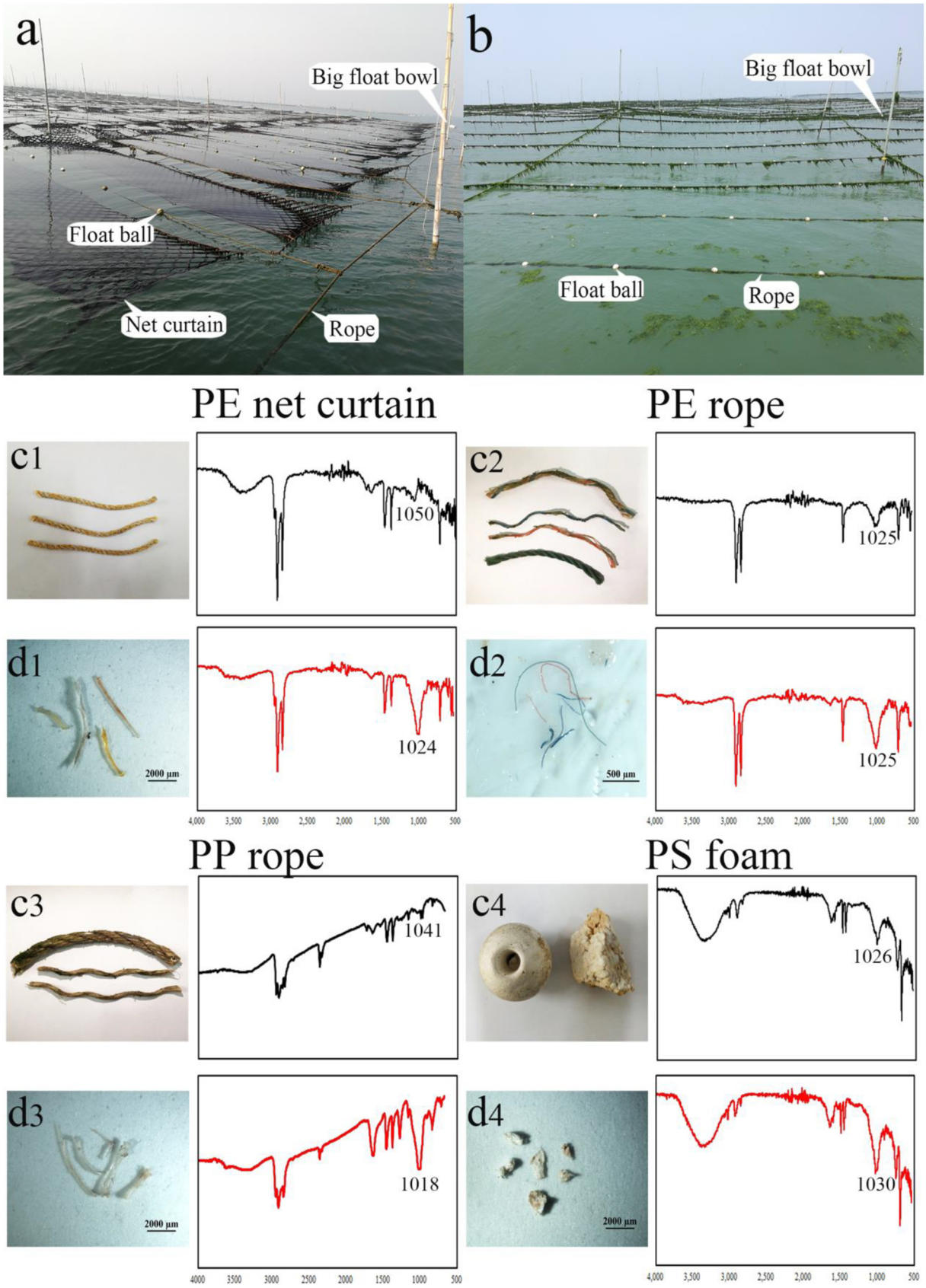

Fig. 3. The culture gears during the culture (a) and non-culture (b) periods, and the FT-IR spectra of the plastics from mariculture gears (c1-c4) and the microplastics from macroalgae (d1-d4).

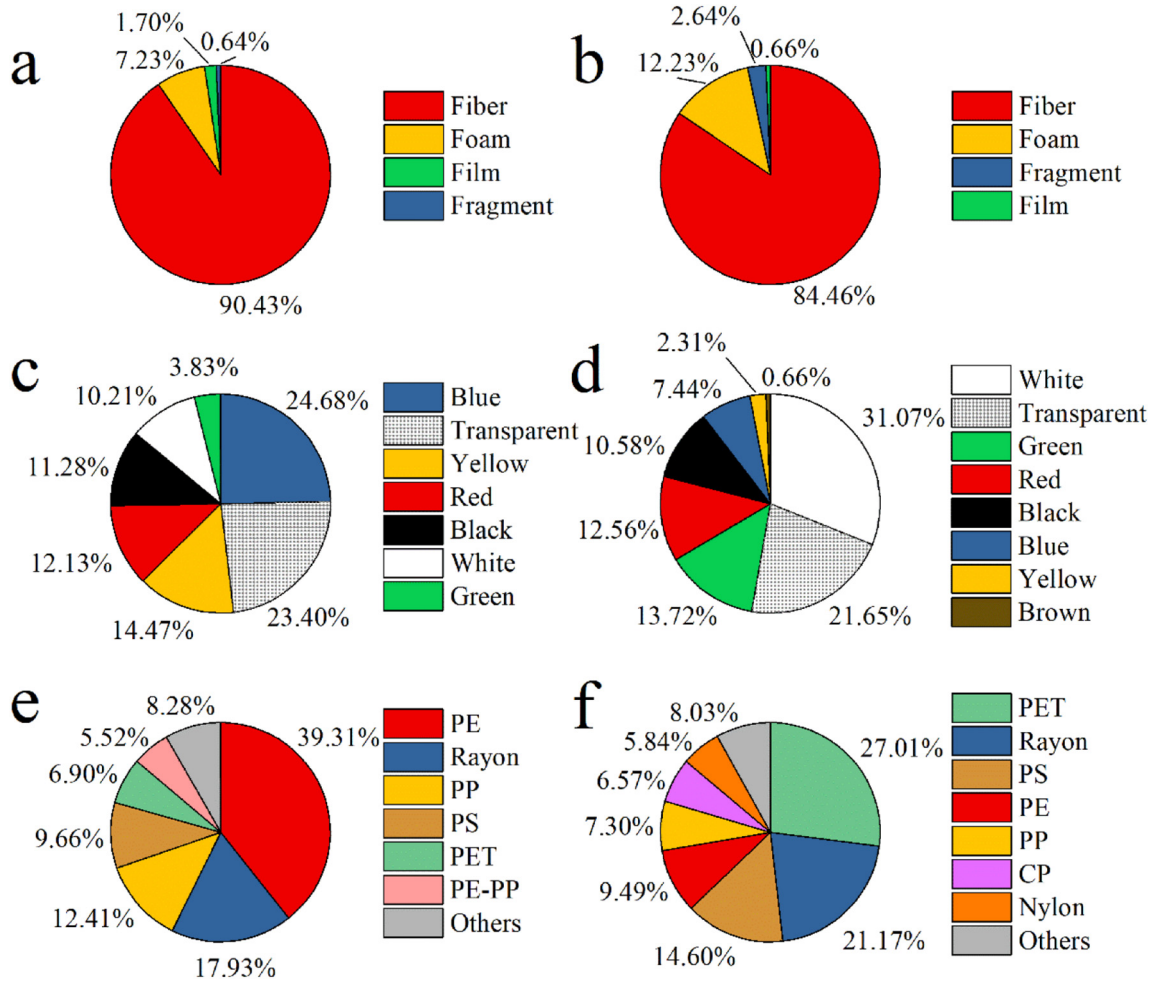

Fig. 4. The shape (a, b), colour (c, d) and material type (e, f) of microplastics on macroalgae during the culture (a, c, e) and non-culture periods (b, d, f). The others involve CP (3.45%), Nylon (2.76%), PAN (1.38%) and PS&PAN&PMMA (0.69%) during the culture period (e), and PAN (3.65%), PE-PP (2.92%) and PMMA (1.46%) during the non-culture period (f).

The size distribution of MPs in different sections was also investigated (Fig. 5). The percentage of MPs < 1 mm in the non-culture period was higher than that in the culture period for each section. In the culture period, S-2 had the highest percentage of MPs < 1 mm while S-1 had the highest percentage of MPs < 1 mm in the non-culture period. On the other hand, macroalgae in each section during the culture period had higher abundance for 1–2 mm MPs compared to those during the non-culture period. When take the culture area as a whole, the abundance of MPs exponentially decreased with the increase of size for both the culture ($y = 826.03611e^{-0.00176x}$, $R^2 = 0.82151$) and non-culture periods ($y = 327.83569e^{-7.00463x}$, $R^2 = 0.98964$).

3.3. MPs in different macroalgae and corresponding seawater

The important data of MPs abundance trapped by different macroalgae were shown (Fig. 6a). *U. prolifera* had the highest MPs abundance (0.19 ± 0.07 particles g^{-1} FW), followed by *P. yezoensis* (0.17 ± 0.08 particles g^{-1} FW) and *S. horneri* (0.14 ± 0.11 particles g^{-1} FW), although the differences among these three species were not statistically different. *U. pertusa* had the lowest MPs abundance (0.06 ± 0.04 particles g^{-1} FW) among the six species investigated here. The MPs abundance (1.04 ± 0.32 particles L^{-1}) in seawater where plants *P. yezoensis* was commonly lower than those (1.63 ± 0.54–1.92 ± 0.75 particles L^{-1}) where plants other macroalgae (Fig. 6b). In terms of the ratio of MPs abundance on macroalgae (A_M) to that in seawater (A_S) (Fig. 6c), *P. yezoensis* had the highest value (160.08 ± 61.86), followed by *U. prolifera* (116.65 ± 43.76) and *S. horneri* (93.75 ± 72.47), and *U. pinnatifida* (57.18 ± 4.04) and *U. pertusa* (34.13 ± 11.96) had the lowest values, implying that *P. yezoensis* has the strongest capacity of trapping MPs among the macroalgae investigated in Haizhou Bay. The shape and size distributions of MPs for each species of macroalgae were analyzed (Figs. S2 & S3). Fiber MPs was the dominant form and smaller MPs accounted for higher proportion for each macroalgae species.

Different colours and shapes of MP trapped on different macroalgae were observed under stereo microscope (Fig. 7). Adherence was the most important strategy that traps MPs for all species of macroalgae (Fig. 7a, b, c). Furthermore, film and foam were twined by branches of *S. horneri* and *Chladophora* (Fig. 7d, e). More interestingly, some MPs of foam were wrapped by *U. prolifera* as part of air sac to afloat (Fig. 7f).

4. Discussion

4.1. MPs trapping by macroalgae

There are a number of studies showing that MPs can be ingested by marine animals, including zooplankton, shellfish, fish, mammals and seabirds, via direct ingestion or food web transfer (Provencher et al., 2018; Qu et al., 2018; Sun et al., 2018; Feng et al., 2019; Nelms et al., 2019). However, little is known about the MPs pollution status in marine primary producers, particularly macroalgae. This study demonstrates that macroalgae can also trap MPs. Compared to marine

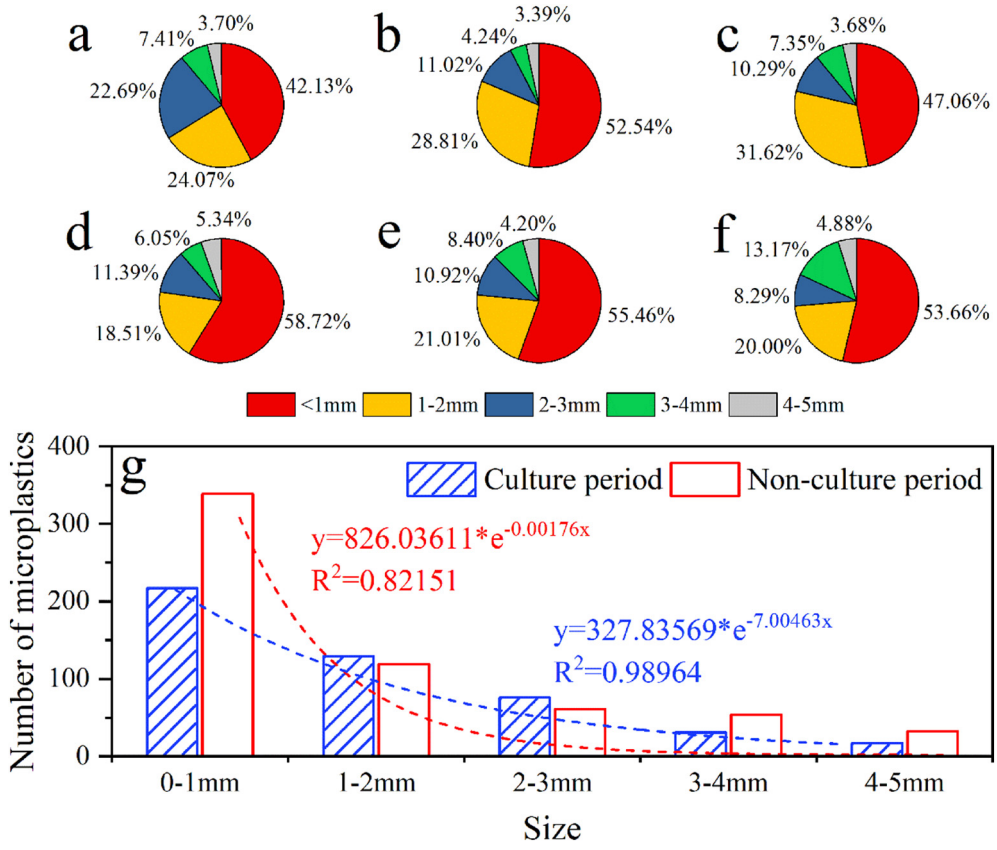

Fig. 5. Size distribution of microplastics on macroalgae in sections of S-1 (a, d) S-2 (b, e), S-3 (c, f) and the whole sea area (g) during the culture (a, b, c) and non-culture periods (d, e, f).

animals that usually trap MPs only in digestive tissues (Law, 2017; Wang et al., 2019b), the whole-thallus of macroalgae can trap MPs (Fig. 7). The skeletal polysaccharides in cell wall of macroalgae could contribute to the adherence of MPs (Frei and Preston, 1964; Xu, 2014; Lagarde et al., 2016). Meanwhile, macroalgae were found to have different capacity to trap MPs in the present study. This could be related to morphology and surface stickiness of macroalgae. Among the macroalgae investigated in this study, *P. yezoensis* showed the strong capacity to trap MPs, which has a sheet-like thallus and could easily adsorb MPs (Li et al., 2020). In addition, their thalli are very soft, making it easy to fold with waves and winds and to twine and wrap MPs. *U. prolifera* ranks second among the macroalgae. Thalli of *U. prolifera* are tubular and it is easy for fiber MPs to twine. In addition, *U. prolifera* has air sac that can wrap different shapes of MPs in it. The MPs abundance in *P. yesoensis* was 0.17 ± 0.08 particles g^{-1} FW (1.53 ± 0.72 particles g^{-1} DW), which is comparable to Li et al.'s (2020) study. The MPs in seawater in this study ranged 0.46–2.82 particles L^{-1} with an average of 1.45 ± 0.59 particles L^{-1}, which was higher than that (0.55 ± 0.28 particles L^{-1}) in North Yellow Sea (Zhu et al., 2018). This difference may be due to different intensities of plastics release from mariculture and inputs from local lands.

Apart from the loads of input from land, human activity in the seas is considered as another source for marine MPs. Fishing gear and marine vessels can lead to the increase of MPs in oceans (Cole et al., 2011; Carney Almroth and Eggert, 2019). Chen et al. (2018) investigated the MPs pollution from mariculture (mainly for marine animals) in a semi-enclosed narrow bay where few seawater exchange with open seas occurs and found that mariculture-derived MPs made up of approximately 55.7% and 36.8% of the MPs in seawater and sediment, respectively. However, little is known the MPs pollution of macroalgae culture in open seas and the interaction MPs pollution of macroalgae culture and macroalgae. The present study shows that MPs fiber made of PE and PP could be released from culture net curtains and ropes and then be trapped by macroalgae, as the colour, shape and material of MPs between culture gears and *P. yezoensis* are very similar. The deeper peaks between 1010 and 1050 cm^{-1} in the spectra of MPs in macroalgae indicates strengthening of the C—O stretching vibration due to weathering effects (Żenkiewicz et al., 2003; Chen et al., 2018). The increased proportion of PE and PP in *P. yezoensis* during culture period confirms MPs pollution of culture because human activity during culture period can stimulate the release of plastics from culture gears. Our study indicates that macroalgae culture can be a source of MPs and the MPs released from culture gears can be effectively trapped by macroalgae attached to culture gears.

As shown in the present study, culture gears, including rope, net curtain, big floating bowl and floating ball, are all made of plastics. The total plastics used in culture gears in Haizhou Bay was about 45, 809 tons in 2018 (Table 2), with an average usage amount of 1.25 tons/ha. These culture gears in a sea area of 58, 589 ha (responding to 36, 618 ha area of net curtain for *P. yezoensis* culture) released about 1, 037 tons plastics into the seas annually. This means these culture gears can increase calculated local MPs concentration of 1.77 mg L^{-1} in 1 m-deep water column. This is a great number since the MPs concentration in surface seawater usually does not surpass 0.1 mg L^{-1} (Goldstein et al., 2012; Kooi et al., 2016; Frère et al., 2017). It is worth noting that although culture gear is a source of MPs, the MPs abundance in seawater during culture period was lower than that during non-culture period. It could be due to the massive trapping of MPs by *P. yezoensis*. For instance, the production of *P. yezoensis* in Haizhou Bay in 2018 achieved 41, 860 tons (CFSY, 2019). Based on the present study, it indicates that about 70 billion particles of MPs in seawater were trapped by *P. yezoensis*. In addition, only net curtain was removed during the non-culture period

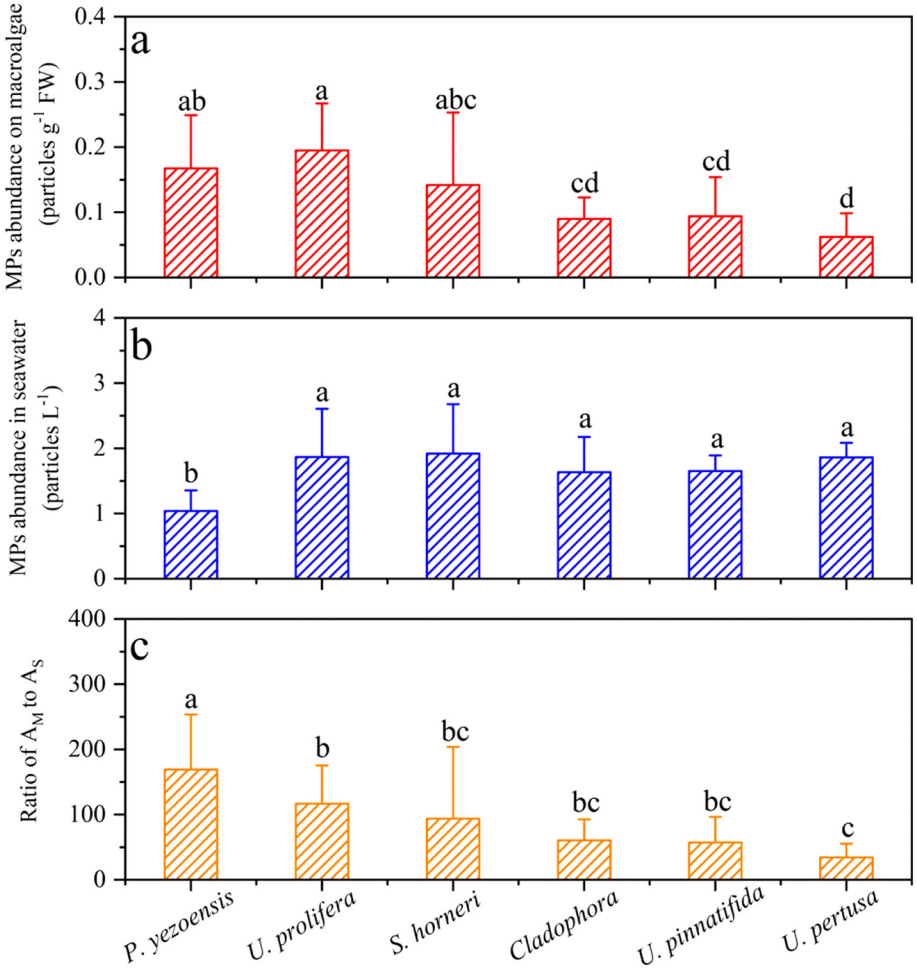

Fig. 6. Microplastics abundance on different macroalgae (a), in seawater around macroalgae (b) and ratio of MPs abundance on macroalgae (A_M) to that in seawater (A_S) (c). Different letters above the bars indicate significant differences among species or seawaters (One-way ANOVA, $P < .05$).

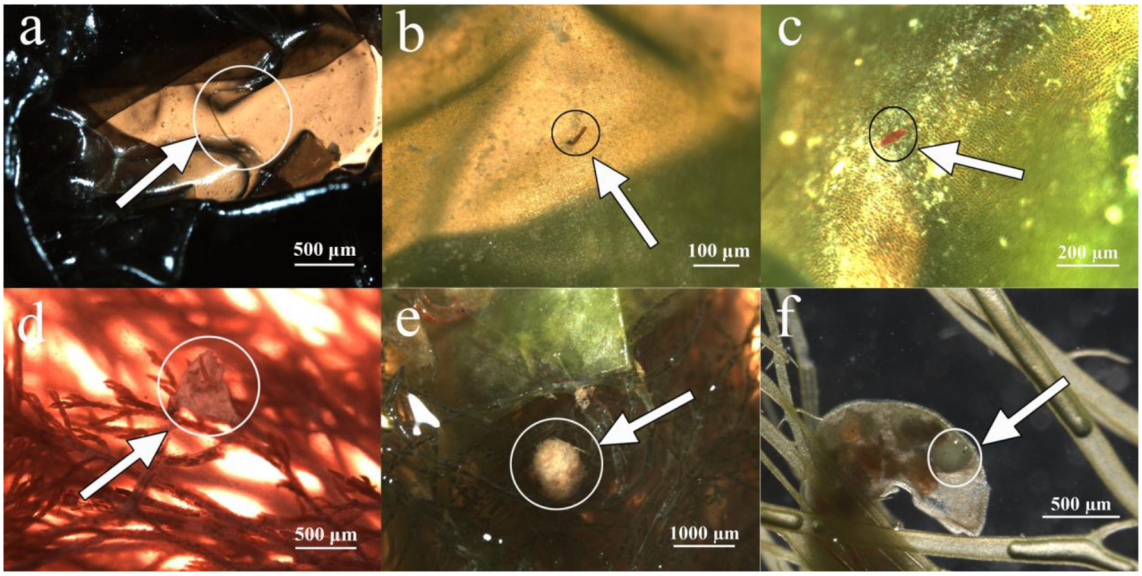

Fig. 7. The trapping approaches of microplastics on six species of macroalgae: (a) blue fiber adherence to the surface of *P. yezoensis*, (b) black fiber adherence to the surface of *U. pinnatifida*, (c) red fiber adherence to the surface of *U. pertusa*, (d) white film twined by *S. horneri*, (e) white foam twined by *Chladophora*, (f) white foam wrapped in *U. prolifera*. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

while other plastic gears were still in the sea, making the gap of plastic release from culture gears between the culture and non-culture periods very small.

4.2. Spatio-temporal variation of MPs pollution on macroalgae

Macroalgae in Section 1 had higher MPs abundance compared to Sections 2 and 3, which could be attributed to the input from land because Section 1 is closest to the coast; while Sections 2 and 3 were less affected by terrestrial input as culture gears and macroalgae could reduce water velocity and dampen wave motion (Løvås and Tørum, 2001; Hendriks et al., 2010). This presumption was supported by the spatial change of salinity and pH where Section 1 had lower salinity and pH compared to Sections 2 and 3 (Table 1). In addition, the negative correlation between seawater salinity and MPs on macroalgae (Fig. 2c) also indicates the impacts of land-derived MP. Compared to the non-culture period, MPs abundance in macroalgae during the culture period was higher for Sections 1 and 2 but not for Section 3. Section 3 is closest to the open sea and easier to exchange with pelagic waters. Given lower MPs abundance in pelagic waters (Cai et al., 2018), this exchange may dilute the MP released from culture gears, leading to the insignificant difference in MPs abundance of macroalgae between the culture and non-culture periods. In addition, there were more PET and Rayon MPs in macroalgae during the non-culture period compared to the culture period, which could be attributed to more land-derived MPs during the non-culture period since it was in the wet season (Cheung et al., 2016; Peng et al., 2018).

Currently marine animals are examined as potential biomonitors for MPs pollution in seawater, referring to seabird, turtle, mussel and fish (Bonanno and Orlando-Bonaca, 2018; Duncan et al., 2019; Su et al., 2019). However, it is difficult to quantitatively identify the correlation of MPs between animal ingested and those in the environment due to long and frequent migration of animals. In addition, animals may selectively ingest MPs, making them poor bioindicators of MP pollution in the environment (Costa et al., 2019; Ward et al., 2019). On the other hand, macroalgae usually live in a certain area via attaching to substrates and they can trap MPs non-selectively via diverse approaches. From this perspective, macroalgae may be a better biomonitor for plastics pollution in seas.

4.3. Risks of MPs in/on edible macroalgae

The red macroalgae genus *Pyropia*, has been an important marine crop in East and Southeast Asia for thousands of years (Bito et al., 2017). Up to 1, 353 thousand tons of *Pyropia* (fresh weight) were cultivated and harvested in 2016 and the total value is about US$ 2.3 billion (FAO, 2018). Among the genus *Pyropia*, *P. yezoensis* is the world's most lucrative marine crop due to its high nutrition value and delicious flavor (Bito et al., 2017; Gao et al., 2019). *Undaria pinnatifida* is also one of the most popular macroalgae used for food in Asian countries. The culture and caption productions of *U. pinnatifida* were 2.07 million and 2, 679 tons in 2016, respectively (FAO, 2018). The present study shows that both *P. yezoensis* and *U. pinnatifida* can trap a large number of MPs. These MPs can be transferred to human directly when they are used as food. Marine animals usually accumulate MPs in digestive tissues that are commonly removed before cooking (Bellas et al., 2016; Ory et al., 2017; Duncan et al., 2019), which indicates the MPs pollution from marine animals can be minimized when they are consumed by human. On the other hand, edible macroalgae are usually consumed by human after simple washing. Our previous study demonstrated that washing cannot significantly reduce MPs abundance in *P. yezoensis* (Li et al., 2020) and thus the MPs adherence to surface of edible macroalgae and wrapped inside these macroalgae may be largely transferred more to human than marine animals to a large extent. This may produce a high potential risk of human health through dietary pollutant exposure, particularly when MPs carry heavy metals and persistent organic pollutants since about 70 billion particles of MPs in seawater could be trapped by *P. yezoensis* based on the production of *P. yezoensis* in Haizhou Bay in 2018 (CFSY, 2019).

4.4. Impacts of MPs in/on non-edible macroalgae

For those macroalgae which are not extensively consumed by human for now, such as *U. prolifera, S. horneri, Cladophora, U. pinnatifida* and *U. pertusa,* they can be consumed by fish and shellfish and MPs in macroalgae can be transferred via food webs (Gutow et al., 2015; Carr et al., 2018). Furthermore, *U. prolifera* and *S. horneri* are two main bloom-forming species that form green tides and golden tides respectively (Smetacek and Zingone, 2013; Gao et al., 2016; Liu et al., 2018). Both *U. prolifera* and *S. horneri* have air sacs or bladders that enable them to float in the surface seawater and expand to a large scale with ocean currents and waves (Cui et al., 2018; Ito et al., 2019). The MPs in *U. prolifera* and *S. horneri* can be transported with the development and movement of algal blooms. Meanwhile, MPs in *U. prolifera* and *S. horneri* can be kept in surface water during algal bloom and may sink to bottom of seas with dead thalli when algal blooms end. Therefore, the spatio-temporal distribution of MPs in the oceans can be altered when they are trapped by these bloom-forming macroalgae. Green and golden tides occur in many areas in the world and the outbreak of them is in a rising trend (Smetacek and Zingone, 2013; Gao et al., 2017). Accordingly, the strong trapping capacity of macroalgae for MPs would impose a substantial impact on MPs fate in the oceans.

5. Conclusions

For the first time, our study investigated the effect of macroalgae culture on macroalgae in terms of MPs pollution. The culture gears for *P. yezoensis* in Haizhou Bay can release about 1, 037 tons plastics into the seas annually. MPs released from nori culture can be trapped by nori itself, resulting in a high abundance in *P. yezoensis*. The MPs trapped by edible *P. yezoensis* may be transferred to human and lead to a high potential risk of human health in other pollutants through a popular food of nori in Asian countries. As important primary producers in coastal ecosystem, MPs trapping in macroalgae can also be transmitted to higher trophic levels via food webs and affect the ecosystem in costal environments. To reduce MPs pollution from mariculture, new culture methods or gears should be developed. Turnover culture has been used in some areas of Southern China, which employs fewer plastics compared to raft culture (Fig. S4). New materials should also be explored and used in macriculture gears to alleviate MPs pollution. In addition to MPs pollution from mariculture, this study also supplies helpful information in using macroalgae as bioindicators for MPs pollution in the seas.

CRediT authorship contribution statement

Zhihua Feng: Conceptualization, Methodology, Writing - review & editing, Supervision, Funding acquisition. **Tao Zhang:** Conceptualization, Methodology, Writing - original draft. **Jiaxuan Wang:** Investigation, Formal analysis, Visualization. **Wei Huang:** Methodology. **Rui Wang:** Investigation. **Juntian Xu:** Funding acquisition. **Guanghui Fu:** Investigation. **Guang Gao:** Conceptualization, Formal analysis, Writing - original draft, Writing - review & editing.

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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.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.137490.

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