

Contents lists available at ScienceDirect

Journal of Hazardous Materials

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



Microplastics in the commercial seaweed nori

Qipei Li^a, Zhihua Feng^b, Tao Zhang^b, Cuizhu Ma^a, Huahong Shi^{a,*}

^a State Key Laboratory of Estuarine and Coastal Research, East China Normal University, Shanghai 200241, China
^b Jiangsu Key Laboratory of Marine Bioresources and Environment, Jiangsu Ocean University, Lianyungang 222005, China

GRAPHICAL ABSTRACT



ARTICLE INFO

Editor: R Teresa *Keywords:* Commercial products Microplastics Nori Processing stages Seaweed capsule Microplastics were ubiquitous but low in nori

ABSTRACT

Microplastics have been reported to attach to the marine macroalgae which act as the vector for microplastic transfer in the marine food web. In this study, the edible seaweed nori (*Pyropia* spp.) was chosen as a target species. The microplastic contaminant situations in nori were analyzed in both its final commercial products and the intermediate products across different processing stages. The abundance of microplastics ranged from 0.9 to 3.0 items/g (dw) among 24 brands of commercially packaged nori samples. With the development of nori processing stages, an enlarged size fraction of greater microplastics (1–5 mm) was observed. Compared with commercially packaged nori samples, the proportions of polypropylene, polyethylene and poly (ethylene-propylene) copolymers increased, whereas that of polyester decreased in factory-processed nori. Additionally, we further simulated and quantified the number of fluorescent polyester fibers (concentrations: 0, 1000, 5000, 10,000 fibers/L) attach to the algal pieces of *Pyropia yezoensis* under laboratory conditions. The average abundance of microfibers on the nori was positively and quantitatively related to their abundances in seawater (p < 0.01). To our best knowledge, this is the first work that shows the prevalence of microplastics in the commercial seaweed nori and relates to their potential sources during the processing phase.

1. Introduction

Microplastics (MPs; defined as < 5 mm in the largest dimension) as an emerging environmental pollutant have been of particular concern around the world (Law and Thompson, 2014). In recent years, researchers have found that MPs were ubiquitously distributed in various environments, including sea surface water, water columns, marine sediments, lakes, rivers, polar glaciers, the soil and the atmosphere (Browne et al., 2011; Van Cauwenberghe et al., 2013; Cozar et al., 2014; Eriksen et al., 2013, 2014; Lebreton et al., 2017; Lusher et al.,

* Corresponding author. *E-mail address:* hhshi@des.ecnu.edu.cn (H. Shi).

https://doi.org/10.1016/j.jhazmat.2020.122060 Received 30 October 2019; Received in revised form 25 December 2019; Accepted 8 January 2020 Available online 09 January 2020

0304-3894/ ${\ensuremath{\mathbb C}}$ 2020 Elsevier B.V. All rights reserved.



2015a; Kim and An, 2019; Liu et al., 2019). After MPs are attached by algae or microorganisms in the ocean to form biofilms, their densities increase and gradually sink from the sea surface, thereby causing MPs to suspend in the water or settle down in the seabed. There have been several reports showing that MPs can migrate downward from the surface water in the form of feces and marine snow after being ingested by organisms (Clark et al., 2016; Karlsson et al., 2017; Katija et al., 2017; Porter et al., 2018).

When organisms contaminated by MPs are ingested by other organisms, MPs can also be transferred and accumulated between organisms of different trophic levels in the food web (Lusher et al., 2015b; Toussaint et al., 2019; Wesch et al., 2016). To date, more than 690 marine species, including bivalves, crustaceans, fishes, sea mammals, and seabirds, have been testified to be contaminated by MPs (Carbery et al., 2018). However, very limited field studies have been focused on investigating microplastic pollution in phytoplankton or macroalgae.

As primary producers in the marine ecosystem, seaweeds provide food and habitats for their consumers and many other associated organisms (Taylor and Cole, 1994). Nevertheless, interactions of MPs with seaweeds and their fate in the marine food web are yet not well understood. So far, one laboratory study has proved that the periwinkle *Littorina littorea*, which feeds on the seaweed *Fucus vesiculosus*, can ingest both MPs and non-MPs contaminated algal food without any preference (Gutow et al., 2016). Therefore, seaweeds can adsorb MPs and facilitate their transfer to organisms of higher trophic levels.

Furthermore, some species of seaweeds, such as nori *Pyropia* spp., are also important ingredients of seafood and have close relationships with human health. In the year of 2010, there were 19 million tons of global seaweed production with an estimated market value of 5.7 billion USD (FAO (Food and Agriculture Organisation of the United Nations), 2010). China, the largest seaweed-producing country in the world, accounted for about 60% of farmed seaweed yield by quantity and 45% by value worldwide (FAO (Food and Agriculture Organisation of the United Nations), 2011). The red seaweed nori *Pyropia* spp. is an edible vegetable and easily processed, thereby it has a large number of farming areas along the coastal waters of China. Accordingly, if the water environments that used for nori cultivation have been reported highly polluted by MPs, then MPs may contaminate and retain in this commercially important seafood species nori even after they are processed and packaged.

In the present study, we hypothesize that the contamination of MPs may be ubiquitous in marketed nori products and MPs may occur and change in nori during its processing phase. Thus, to test these hypotheses, we analyzed the contaminant situations of MPs in both nori's final commercial products and the intermediate products across different processing stages. Additionally, we simulated and quantified the number of adherent fluorescent polyester fibers to algal pieces of *Pyropia yezoensis* under laboratory conditions to establish the relationship between microplastic levels in nori and the seawater. Together, these results will be used to determine the possible sources of MPs in nori and assess the potential risks to human health.

2. Materials and methods

2.1. Collection of nori

Twenty-four brands of commercial dried nori were purchased from local markets in China in January and February 2019. These commercial products came from eight provinces and one municipality along the east coastal zone of China (Fujian Province: nine brands; Shandong Province: four brands; Hainan Province: three brands; Liaoning Province and Shanghai Municipality: two brands each; Jiangsu, Zhejiang, Guangdong, and Guangxi provinces: one brand each). A pack of dried nori with a range of weight from 30 to 100 g was selected for each brand. In addition, fresh nori (*Pyropia* spp.) was collected from three nori farm sites (NF₁, NF₂, NF₃) in the Yellow Sea in February 2019 (Fig. S1). Their corresponding washed and dried products were obtained sequentially from three different nori processing factories located in Lianyungang City, Jiangsu Province, China.

2.2. Quality control of experiments

To prevent the potential cross-contamination, all of the liquids (artificial seawater, hydrogen peroxide, and saline solution) were filtered through glass fiber filters (Whatman GF/B CAT No. 1821-047, pore size = 1 μ m, filter diameter = 47 mm) before use. All laboratory glasswares and tools were cleaned with 70% ethanol and rinsed three times with MilliQ water (Millipore, Bedford, USA). The samples in clean Petri dishes were immediately covered with aluminum foil or glass lid when they were not in use. Non-latex nitrile gloves and cotton laboratory coat were worn during the whole experimental process.

2.3. Measurement of the psychrometric ratio of nori

Clean and dried beakers were covered with aluminum foil and weighed (W_0). Approximately 100 g of wet nori (unwashed and washed) were transferred to the beakers, respectively. Then the beakers with wet nori were weighed (W_1), placed and dehydrated in a convective oven (DHG-9123A, Jinghong Co., Ltd., Shanghai, China) at 105 °C for 8 h. After drying, the beakers with dried nori were weighed (W_2) again. The weight of each sample was measured three times. The psychrometric ratio of nori was calculated by the following formula:

The psychrometric ratio = $\frac{W_1 - W_0}{W_2 - W_0} \times 100\%$

2.4. Enzymatic hydrolysis and hydrogen peroxide treatment

The dried algal thalli from commercial packaged nori and laboratory dried ones were picked up with forceps and cut into pieces using dissecting scissors. Approximately 3 g dry weight of small pieces of nori algal sheet was placed into a 1 L glass bottle, with three replicates were taken separately for each commercial brand or processing stage. Blank controls without samples were simultaneously performed to assess air contamination. Approximately 100 mL of cellulase solution (0.1%, v/v) was added to each bottle to decompose the cytoderm of algal cells. The pH was adjusted to 4.5-6.0 and the bottles were incubated in an orbital shaker (ZWF-1112, Zhicheng Co., Ltd., Shanghai, China) at 55 °C at 80 rpm for 1 h. Subsequently, $100 \,\mu$ L of alcalase solution (100%, v/v) was added to digest the released protein after decomposition of the cell wall. The pH of the solution was elevated to 6.0-7.0 and the bottles were placed in the shaker at 55 °C at 80 rpm for another 1 h. After twostep enzymolysis, approximately 400 mL of H₂O₂ solution (30%, v/v) was added to each bottle. Then the bottles were set in the shaker at 65 °C at 80 rpm for 72 h until digestion is completed.

2.5. Floatation and isolation of MPs

The floatation and isolation of MPs were performed as previously described (Li et al., 2016). Briefly, approximately 500 mL of filtered saturated solution of NaCl was added to separate MPs from the organic matter in the dissolved liquid. Then the cocktail of liquid was mixed and held overnight. After floatation, the supernatants of liquid were directly filtered over a nitrocellulose membrane filter (Millipore SMWP04700, pore size $=5 \,\mu$ m, filter diameter $= 47 \,\text{mm}$) by a vacuuming system. After filtering the liquid, the membrane filters were collected in clean Petri dishes for further microplastic verification.

2.6. Visual identification and validation of MPs

Digital images of plastic-like particles on the membrane filters were taken under a stereo optical microscope (Zeiss Discovery V8, Göttingen, Germany) equipped with an AxioCam Icc3 camera. Then a number of suspected plastic particles were arbitrarily selected for verification of MPs using a micro-Fourier Transform Infrared (μ -FT-IR) spectroscope. The infrared spectra of samples were recorded on Nicolet iN10 MX (Thermo Scientific, USA). The OMNIC operating system in the transmission mode was used to obtain data with a 4 cm⁻¹ resolution and 32 scans (Yang et al., 2015). The polymer type of samples was verified in comparison with the Thermo Scientific infrared spectra library and our own database of semi-synthetic celluloses. For effective verification of particles, the spectrum matching with similarity index no less than 0.7 was accepted. Of all 693 visually identified particles, 328 particles (47.3%) were instrumental verified (Supplementary Table S1). Such a validation rate and a number of analyzed particles meet the general requirements (close to 50% validation rate with a minimum of 100 particles) in the field of microplastic research (Hermsen et al., 2018).

2.7. Adherence of fluorescent MPs to nori in the laboratory

To explore how the seaweed gets MPs in the field and if the MPs can adhere to the surface of nori, a laboratory experiment was conducted. The fluorescent polyester fibers (Ex: 341 nm, Em: 530 nm) were produced by blending the polyethylene terephthalate (PET), luminescent powders and coupling agent, which can improve the spinnability and luminesce properties of the fibers. They were used in this experiment and cut into pieces as tiny as possible using dissecting scissors. The approximate size of these microfibers varied from 100 to 2000 μ m. The polymer type of fluorescent fibers was verified by a Thermo Nicolet iS5 FT-IR spectroscope. The aqueous stock suspension of microfibers (approximately 100 fibers/mL) was prepared for subsequent exposure experiments with a gridded Sedgewick-Rafter Counting cell (Li et al., 2019).

The laboratory exposure experiment was performed with a modified protocol suggested by Gutow et al. (2016). In brief, small pieces of algal thalli (surface area: $3-6 \text{ cm}^2$) were abscised of *Pyropia vezoensis* that was directly collected from nori farms. The algal pieces were rinsed thrice with MilliQ water to remove epiphytes on the surface of the alga. Then the washed algal piece was individually placed in a glass flask with 40 mL of filtered seawater. The seawater was supplemented with fluorescent microfibers from a serial dilution of aqueous stock to the following concentrations: 0, 1000, 5000, 10,000 fibers/L. Each concentration treatment comprised 6 replicates. The algal pieces were whirled on an orbital shaker at 9 °C at 60 rpm for 2 h in seawater containing fluorescent microfibers. Then the flasks of each concentration treatment were halted and sampled for further microplastic analysis. In the end, all algal pieces from exposure flasks were picked out, tiled and dried on slides for 1 h. The numbers of fluorescent microfibers on the upper layer of the algal piece were quantified with a fluorescence microscope (Olympus BX53, Tokyo, Japan). The length of microfibers and the surface area of the tiled alga were measured by an image processing Image J software (NIH, Bethesda, MD, USA).

2.8. Statistical analysis

Normal distribution of data and homogeneity of variances were examined using the Shapiro-Wilk and Levene's test, respectively (SPSS 22, SPSS Inc., Chicago, Illinois, USA). The Kruskal-Wallis ANOVA analysis was used to determine whether there was a significant difference in microplastic abundances for more than two commercial brands of nori samples. Two-way ANOVA test was used to analyze the effect of source or processing on the abundance of MPs in nori samples. Statistical significance was accepted at p < 0.05 and values were presented as means \pm standard deviation. A linear regression analysis was applied to determine the significant correlation between the abundances of fluorescent microfibers in seawater and on the surface of the algae. The figures and maps were plotted and generated using software Origin pro 9 (Origin Lab Co., Northampton, MA, USA) and



Fig. 1. Abundance of MPs in nori samples with different commercial brands from the markets. Each bar represents mean \pm SD of three replicates (n = 3). Letters above each bar indicate the result of multiple comparisons of microplastic abundance; the bars sharing the same letter are not statistically significant.

Surfer 16 (Golden Software, Inc., Golden, Colorado, USA).

3. Results

3.1. Characteristics of MPs in commercially packaged nori

In the investigation of MPs occurrence in commercially packaged nori, all selected brands excluding one brand (B₂) were identified to be contaminated by MPs. As shown in Fig. 1, the abundance of MPs detected from commercially packaged samples ranged from 0.9 to 3.0 (average: 1.8 ± 0.7) items/g. dry weight (dw). Compared to these detected MPs found in commercially packaged nori, the MPs for the blank control (0.1 ± 0.2 items/g. dw) is negligible. However, no significant differences in microplastic abundances were found among different commercial brands (Kruskal-Wallis test, p = 0.060).

The size, shape, and color distributions of MPs in commercially packaged nori are shown in Fig. S2. The size of MPs in commercially packaged samples ranged from 0.11 to 4.97 mm, and the median size was 1.13 mm. MPs with sizes greater than 1.0 mm (25–90%, average: 57.8%) were more recurrently detected than other size fractions (Fig. S2A). Fiber was the predominant shape of MPs (50–100%, average: 85.2%), followed by fragment, film, and pellet (Fig. S2B). The color categories of MPs were classified according to the methods of Feng et al. (2019). Blue-green colored MPs (10–76.5%, average: 41.4%) were more frequently observed than other color classes (Fig. S2C). As shown in Fig. S3, several representative morphotypes of MPs in commercially packaged nori were photographed and further verified for polymer types.

Of the 227 effectively verified particles, 89 plastic items (39.2%) being affiliated with 11 polymer types were verified from commercially packaged nori samples (Supplementary Table S2). Polyester fibers were the most dominant detected MPs (18.9%), followed by rayon (6.6%), polypropylene (4.0%), polyamide (1.8%) and cellophane fibers (1.8%). In addition, non-plastic particles, including cotton and naturally occurring cellulosic fibers, accounted for 60.8% of total particles measured.

3.2. Characteristics of MPs in factory-processed nori

The psychrometric ratios of unwashed nori were 11.1 \pm 0.2%, 16.2 \pm 2.9%, and 11.2 \pm 0.4%, and those of washed nori were 9.2 \pm 0.2%, 3.0 \pm 0.4%, and 3.5 \pm 0.3% for sources from NF₁, NF₂,



Fig. 2. Abundance of MPs in nori samples across different processing stages from source NF₁ (A), NF₂ (B), and NF₃ (C), respectively. Each bar represents mean \pm SD of three replicates (n = 3). Letters above each bar indicate the result of multiple comparisons of microplastic abundance; the bars sharing the same letter are not statistically significant.

and NF₃, respectively. After conversion to the same term of dry weight, the overall abundance of MPs in factory-processed nori ranged from 1.0–2.8 (average: 1.8 ± 0.6) items/g. dw (Fig. 2). Two-way ANOVA indicated that the source (F_{2,18} = 4.551, p = 0.025) affected the MPs abundances significantly, whereas the processing (F_{2,18} = 2.807, p = 0.087) didn't. Specifically, the Post-hoc LSD test suggested that the average abundance of MPs in nori samples from NF₃ farm was significantly lower than samples from NF₁ or NF₂ farms.

The size, shape, and color distributions of MPs in factory-processed nori samples are shown in Fig. S4. The size of MPs in factory-processed samples ranged from 0.07 to 4.74 mm, and the median size was 0.85 mm. The fraction of MPs sizes greater than 1.0 mm (33.2–53.3%, average: 43.4%) increased along with the production process (Fig. S4A). The primary shapes of MPs were fibers (57.1–74.9%, average: 64.8%), followed by fragments, films, and pellets (Fig. S4B). Similar to what was found in commercially packaged nori, blue-green colored MPs (43.9–53.9%, average: 48.1%) were the most prevalent class in factory-processed samples (Fig. S4C).

Of the 86 effectively verified particles, 62 plastic items (72.1%) belonging to 10 polymer types were validated from factory-processed

nori samples (Supplementary Table S2). Among them, polypropylene fibers were the most commonly identified MPs (16.3%), followed by rayon (10.5%) and polyester fibers (9.3%). To track the origin of polypropylene MPs detected in potential manufacturing or industrial uses, different sizes of polypropylene fibers were analyzed in nori samples from a farm site (Fig. 3A), unprocessed (Fig. 3B) and dried products (Fig. 3C) from a processing factory, respectively.

3.3. Adherence and quantification of MPs to nori in the laboratory

Upon UV light activation on the surface of algal pieces of *P. yezoensis* (Ex: 341 nm, Em: 530 nm), no fibers were observed in the control whereas fluorescent polyester microfibers were readily visible in the exposure treatments (Fig. 4A and B). The length of the fluorescent microfibers measured in this experiment ranged from 85 to 2123 (average: 656 ± 384) µm. When algal pieces of *P. yezoensis* were contaminated by fluorescent microfibers, the average abundance of fibers on the nori was positively related to the fiber number concentrations in seawater (Linear Regression, $F_{1,22} = 54.818$, p < 0.01). Significantly lower numbers of adherent fibers on algal pieces were counted at the lowest concentration compared to the other two higher concentrations (Krusal-Wallis test, p < 0.05) (Fig. 4C). Thus, these results suggest that microplastic abundances attached to the surface of nori are positively and quantitatively related to microplastic concentrations in its surrounding waters.

4. Discussion

4.1. Microplastic pollution in commercially packaged nori

In the current study, we investigated microplastic pollution among 24 selected brands of commercially packaged nori samples. MPs were found in 23 out of 24 analyzed samples. The high frequency of microplastic detection rate (95.8%) is much greater than that reported in canned sprats and sardines (20%), but it is close to that reported in commercial salts (94.1%) (Karami et al., 2018, 2017). These discrepancies may due to different analytical methods, food sources and food processing procedures. The abundance of MPs in nori ranged from 0.9 to 3.0 (average: 1.8 \pm 0.7) items/g. dw, and is comparable with those previously found in commercial bivalves (average: 2.5 ± 1.5) from the local markets of China, and live and processed mussels (0.9-1.4) sold in the supermarkets of UK (Li et al., 2015, 2018). However, on basis of the results of the available studies in other types of regularly consumed food, such as honey, salt and sugar, the abundances of MPs were not reported in a unified measuring unit in these samples (Hermsen et al., 2018; Karami et al., 2017; Liebezeit and Liebezeit, 2013, 2015; Muhlschlegel et al., 2017; Kosuth et al., 2018). Therefore, it is still hard to compare microplastic contamination levels among these food items without bias.

Since plastic waste has been suggested being classified as a hazardous substance, the occurrence of MPs in seafood poses potential health risks to humans by seafood consumption (Rochman et al., 2013; Alexander et al., 2016). Regarding toxicity of MPs, several preliminary medical studies have demonstrated adverse effects of microplastic exposures, including size-dependent effect of plastic particles, transfer of additives and adsorbed chemical contaminants, disturbance of the gut microbiome, induced oxidative stress and enhanced inflammatory response (Frohlich et al., 2009; des Rieux et al., 2005; Eldridge et al., 1989; Volkheimer, 1975; Brown et al., 2001; Hussain et al., 2001). Wright and Kelly (2017) predicted that particle size, shape, chemical composition, hydrophobicity, surface charge, and functional groups are influencing factors to determine the uptake of MPs by humans. However, without further investigation, standardized methods and quantification of MPs being translocated through popular seafood species, it is hard to conduct an accurate exposure assessment on the trophic transfer of MPs to higher levels of organisms, and consequently humans.



Fig. 3. Analysis of MPs in nori samples with optical microscopy and μ -FT-IR spectroscopy. Nori samples in the first column (A₁, B₁, C₁) were photographed from a nori farm site, a nori processing factory, and a dried nori product, respectively. Optical photographs in the second column (A₂, B₂, C₂) showed the plastic particles isolated from different sources to their left ones, and the third column (A₃, B₃, C₃) displayed FT-IR spectra of the corresponding particles to their left ones. These particles were all identified as polypropylene in the top left corners of panels.

Therefore, more research is needed to further investigate factors influencing microplastic loads by these seafood species, their bioaccumulation and biomagnification factors, as well as their trophic interactions in the marine food web. With the continuous increase of MPs in the environment, this field of research requires urgent and systematic investigation to inform risk assessments of the impact of MPs on human health.

4.2. Variations of MPs across different processing stages in nori

Microplastic pollution levels in coastal seafood species are closely related to concentrations of MPs detected in their ambient sediments and waters (Li et al., 2016; Jabeen et al., 2017; Qiu et al., 2015; Mathalon and Hill, 2014; Yu et al., 2016; Zhao et al., 2018). Not surprisingly, we found that the average MPs abundance in unprocessed nori samples differed significantly among different sources. In addition, several laboratory studies have shown that the number of MPs that adhered to the aquatic organisms, such as brown seaweed *Fucus vesiculous* and blue mussel *Mytilus edulis*, was correlated with the concentrations of suspended microparticles in waters (Gutow et al., 2016; Kolandhasamy et al., 2018). On the other hand, processing may play a role in varying the average MPs abundance in nori samples along with

the production process. Interestingly, the abundance of MPs in nori samples from NF_3 farm was continuously increased after the washing and after drying stages compared with those before the washing stage. This is indicated that a relatively higher level of MPs can be introduced during the processing phase although the environmental background microplastic value is low for nori at the farming site.

Furthermore, the fraction of greater sized MPs (1–5 mm) in factoryprocessed nori samples were commonly enlarged across different processing stages. Slightly higher percentages of fibrous and colored MPs were observed in nori samples after the washing stage compared to the other two processing stages. To interpret these variations better, further investigation is needed to understand the bioavailability of MPs to nori in situ, the open and enclosed status of nori washers and dryers, as well as the shedding of clothing that workers worn at different processing stages.

4.3. Potential sources of MPs in nori

The majority of MPs in nori we found were of fibrous shape, which suggests they may come from various sources, including riverine input and sewage effluent from coastal areas, the breakdown of larger plastic debris from local human activities (Browne et al., 2011; Lebreton et al.,



Abundance of microfibers in seawater (items/L)

Fig. 4. Adherence of fluorescent polyester microfibers to nori. Microphotographs of fluorescent polyester microfibers on the surface of algal pieces of *P. yezoensis* in the control (A) and in the exposure treatment (B). Arrows indicate the location of microfibers. Scale bar = 100μ m. Abundance of fluorescent polyester microfibers on algal pieces of *P. yezoensis* that were contaminated at different microfiber number concentrations in seawater (n = 6) (C). The letter above each bar shows the result of multiple comparisons of microfiber densities among different microfiber concentration treatments; the bars sharing the same letter are not statistically significant.

2017; Thompson et al., 2004; Woodall et al., 2014; Mason et al., 2016; Peller et al., 2019; Deng et al., 2019; Su et al., 2019; Severini et al., 2019). The global production of synthetic fibers has been continuously increased since the 1960s, it is therefore that this type of MPs contamination is likely to increase in the marine environment (Thompson et al., 2004; Woodall et al., 2014). According to a global investigation of microplastic pollution along the shorelines, there is evidence that a primary source of MPs is synthetic fibers from washing garments (Browne et al., 2011). Rivers are widely found to represent the major pathway for plastic fibers to enter the aquatic environment (Lebreton et al., 2017). Some point sources, like Wastewater Treatment Plants (WWTPs) effluents and textile industrial discharges, are considered as significant contributors of textile fibers to land-based sources of MPs in the river basins (Mason et al., 2016; Peller et al., 2019; Deng et al., 2019). In addition, the widespread occurrence of synthetic fibers in nori samples analyzed could be related to the intense human activities in the nori's culture and production area, such as the use of plastic lines for

the attaching of seaweed seedlings and from fishing nets. This evidence was also found in similar sea ranching areas for other commercially important species, such as fishes, mussels and oysters (Mathalon and Hill, 2014; Su et al., 2019; Severini et al., 2019). Taken together, these results suggest that there are various potential sources of synthetic microfibers to the aquatic environment and the organisms.

Furthermore, the polymer types of particles detected in various samples are used to trace their origins in potential manufacturing or industrial uses (Li et al., 2018; Mathalon and Hill, 2014; Lusher et al., 2017; Claessens et al., 2011; Desforges et al., 2014). Of these commonly used polymers related to fishery activities, polyethylene, polypropylene, and polyamide are broadly used in textile fabrics and fishing tackle, including ropes, fishing nets and threads (Claessens et al., 2011; Desforges et al., 2014). Polystyrene, polyvinylchloride, and polyethylene terephthalate are mainly used for packaging purposes, e.g. plastic bags, caps, bottles, film, containers, and pipes (Claessens et al., 2011). In our study, different sizes of polypropylene plastic debris were

also identified in factory-processed nori samples (Fig. 3). This is direct evidence to show a connection between plastic materials used in anthropogenic activities and MPs detected in seafood species and their processed products. Nonetheless, polypropylene, polyethylene and poly (ethylene-propylene) copolymers were relatively low or none in total MPs identified in commercially packaged nori samples. It might be caused by specific procedures of nori processing, such as washing and drying, which removed a large number of certain types of plastic polymers from origin samples. In contrast, the proportion of polyester fibers increased in these final commercial products. Previous studies have shown that contamination of textile fibers, which includes MPs, is prevalent in indoor and outdoor air (Dris et al., 2017, 2016). Most recently, Liu et al. (2019) found that polyester (including PET) is the most dominant component in MPs from dust in urban China. Hence, these airborne synthetic fibers were very likely to appear and contribute to the composition of polymers in nori's commercial products during the processing and packaging phases.

In addition to regular synthetic fibers, semi-synthetic cellulosic fibers, such as rayon and cellophane, are also recurrently detected in both biotic and abiotic samples (Lusher et al., 2015a; Woodall et al., 2014; Taylor et al., 2016; Chen et al., 2018; Lusher et al., 2014). These fibers contributed to approximate 30%-60% of suspected MPs analyzed in the above studies, the proportions of which were relatively higher than what we currently found in nori samples. In the present study, we found that the semi-synthetic cellulosic fibers accounted for about 8.4% and 14% of total microparticles measured in commercial and factoryprocessed nori samples, respectively (Supplementary Table S2). The production volume of semi-synthetic cellulosic fibers is higher than all other synthetic organic polymers according to a review of Shen et al. (2012). Also, there have been several reports indicating that these artificial fibers could be the majority of MPs in the marine environment from terrestrial sources (Woodall et al., 2014; Chen et al., 2018). However, it is challenging to distinguish semi-synthetic cellulosic fibers clearly from natural fibers using the traditional spectroscopic methods (Comnea-Stancu et al., 2017; Cai et al., 2019). Therefore, further research is needed to identify their presence and quantify their amount more accurately to better understand their potential sources and fates in various environments.

5. Conclusions

Our results demonstrate that MPs were ubiquitous but low in the commercial seaweed nori. In the investigation of factory-processed nori, the source did significantly influence the abundance of MPs in nori while processing did not. Our findings indicate that the abundance and composition of MPs in nori's final commercial products and the intermediate products are related to microplastic concentration and type in their ambient environments, respectively. Moreover, we need to pay careful attention to discern the potential sources of MPs in these seafood products and assess the human health risks of MPs in future studies.

CRediT authorship contribution statement

Qipei Li: Conceptualization, Methodology, Validation, Data curation, Formal analysis, Investigation, Writing - original draft, Writing review & editing. Zhihua Feng: Investigation, Resources, Writing review & editing. Tao Zhang: Investigation, Writing - review & editing. Cuizhu Ma: Methodology, Writing - review & editing. Huahong Shi: Supervision, Funding acquisition, Writing - review & editing.

Declaration of Competing Interest

The authors declare that there are no known conflicts of interest that could have caused any influence to the work reported in this publication.

Acknowledgments

The authors would like to thank Dr. Gong Jinghua, State Key Laboratory for Modification of Chemical Fibers and Polymer Materials, Donghua University, for his assistance with fluorescent polyester fibers. This work was financially supported through the National Science Foundation of China Grant (No. 41571467) and the National Key Research and Development Grant (No. 2016YFC1402204).

Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.jhazmat.2020.122060.

References

- Alexander, J., Barregard, L., Bignami, M., Ceccatelli, S., Cottrill, B., Dinovi, M., Edler, L., Grasl-Kraupp, B., Hogstrand, C., Hoogenboom, L., Knutsen, H.K., Nebbia, C.S., Oswald, I., Petersen, A., Rogiers, V.M., Rose, M., Roudot, A.C., Schwerdtle, T., Vleminckx, C., Vollmer, G., Wallace, H., Chain, E.P.C.F., 2016. Presence of microplastics and nanoplastics in food, with particular focus on seafood. Efsa J. 14.
- Brown, D.M., Wilson, M.R., MacNee, W., Stone, V., Donaldson, K., 2001. Size-dependent proinflammatory effects of ultrafine polystyrene particles: a role for surface area and oxidative stress in the enhanced activity of ultrafines. Toxicol. Appl. Pharm. 175, 191–199.
- Browne, M.A., Crump, P., Niven, S.J., Teuten, E., Tonkin, A., Galloway, T., Thompson, R., 2011. Accumulation of Microplastic on shorelines woldwide: sources and sinks. Environ. Sci. Technol. 45, 9175–9179.
- Cai, H.W., Du, F.N., Li, L.Y., Li, B.W., Li, J.N., Shi, H.H., 2019. A practical approach based on FT-IR spectroscopy for identification of semi-synthetic and natural celluloses in microplastic investigation. Sci. Total Environ. 669, 692–701.
- Carbery, M., O'Connor, W., Thavamani, P., 2018. Trophic transfer of microplastics and mixed contaminants in the marine food web and implications for human health. Environ. Int. 115, 400–409.
- Chen, M.L., Jin, M., Tao, P.R., Wang, Z., Xie, W.P., Yu, X.B., Wang, K., 2018. Assessment of microplastics derived from mariculture in Xiangshan Bay, China. Environ. Pollut. 242, 1146–1156.
- Claessens, M., De Meester, S., Van Landuyt, L., De Clerck, K., Janssen, C.R., 2011. Occurrence and distribution of microplastics in marine sediments along the Belgian coast. Mar. Pollut. Bull. 62, 2199–2204.
- Clark, J.R., Cole, M., Lindeque, P.K., Fileman, E., Blackford, J., Lewis, C., Lenton, T.M., Galloway, T.S., 2016. Marine microplastic debris: a targeted plan for understanding and quantifying interactions with marine life. Front. Ecol. Environ. 14, 317–324.
- Comnea-Stancu, I.R., Wieland, K., Ramer, G., Schwaighofer, A., Lendl, B., 2017. On the Identification of Rayon/Viscose as a Major Fraction of Microplastics in the Marine Environment: Discrimination between Natural and Manmade Cellulosic Fibers Using Fourier Transform Infrared Spectroscopy. Appl. Spectrosc. 71, 939–950.
- Cozar, A., Echevarria, F., Gonzalez-Gordillo, J.I., Irigoien, X., Ubeda, B., Hernandez-Leon, S., Palma, A.T., Navarro, S., Garcia-de-Lomas, J., Ruiz, A., Fernandez-de-Puelles, M.L., Duarte, C.M., 2014. Plastic debris in the open ocean. Proc. Natl. Acad. Sci. U. S. A. 111, 10239–10244.
- Deng, H., Wei, R., Luo, W.Y., Hu, L.L., Li, B.W., Di, Y.N., Shi, H.H., 2019. Microplastic pollution in water and sediment in a textile industrial area. Environ. Pollut. 258.
- des Rieux, A., Ragnarsson, E.G., Gullberg, E., Preat, V., Schneider, Y.J., Artursson, P., 2005. Transport of nanoparticles across an in vitro model of the human intestinal follicle associated epithelium. Eur. J. Pharm. Sci. 25, 455–465.
- Desforges, J.P.W., Galbraith, M., Dangerfield, N., Ross, P.S., 2014. Widespread distribution of microplastics in subsurface seawater in the NE Pacific Ocean. Mar. Pollut. Bull. 79, 94–99.
- Dris, R., Gasperi, J., Mirande, C., Mandin, C., Guerrouache, M., Langlois, V., Tassin, B., 2017. A first overview of textile fibers, including microplastics, in indoor and outdoor environments. Environ. Pollut. 221, 453–458.
- Dris, R., Gasperi, J., Saad, M., Mirande, C., Tassin, B., 2016. Synthetic fibers in atmospheric fallout: A source of microplastics in the environment? Mar. Pollut. Bull. 104, 290–293.
- Eldridge, J.H., Meulbroek, J.A., Staas, J.K., Tice, T.R., Gilley, R.M., 1989. Vaccine-containing biodegradable microspheres specifically enter the gut-associated lymphoid tissue following oral administration and induce a disseminated mucosal immune response. Adv. Exp. Med. Biol. 251, 191–202.
- Eriksen, M., Mason, S., Wilson, S., Box, C., Zellers, A., Edwards, W., Farley, H., Amato, S., 2013. Microplastic pollution in the surface waters of the Laurentian Great Lakes. Mar. Pollut. Bull. 77, 177–182.
- Eriksen, M., Lebreton, L.C.M., Carson, H.S., Thiel, M., Moore, C.J., Borerro, J.C., Galgani, F., Ryan, P.G., Reisser, J., 2014. Plastic pollution in the world's oceans: more than 5 trillion plastic pieces weighing over 250,000 tons afloat at sea. PLoS One 9.
- FAO (Food and Agriculture Organisation of the United Nations), 2010. The State of the World Fisheries and Aquaculture 2010. Food and Agriculture Organisation of the United Nations, Rome. www.fao.org/docrep/013/i1820e/i1820e00.htm.
- FAO (Food and Agriculture Organisation of the United Nations), 2011. FAO Yearbook -Fisheries and Aquaculture Statistics 2009. Food and Agriculture Organisation of the United Nations, Rome ftp.fao.org/FI/CDrom/CD_yearbook_2009/index.htm.

- 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.
- Frohlich, E., Samberger, C., Kueznik, T., Absenger, M., Roblegg, E., Zimmer, A., Pieber, T.R., 2009. Cytotoxicity of nanoparticles independent from oxidative stress. J. Toxicol. Sci. 34, 363–375.
- Gutow, L., Eckerlebe, A., Gimenez, L., Saborowski, R., 2016. Experimental evaluation of seaweeds as a vector for microplastics into marine food webs. Environ. Sci. Technol. 50, 915–923.
- Hermsen, E., Mintenig, S.M., Besseling, E., Koelmans, A.A., 2018. Quality criteria for the analysis of microplastic in Biota samples: a critical review. Environ. Sci. Technol. 52, 10230–10240.
- Hussain, N., Jaitley, V., Florence, A.T., 2001. Recent advances in the understanding of uptake of microparticulates across the gastrointestinal lymphatics. Adv. Drug Deliver. Rev. 50, 107–142.
- Jabeen, K., Su, L., Li, J.N., Yang, D.Q., Tong, C.F., Mu, J.L., Shi, H.H., 2017. Microplastics and mesoplastics in fish from coastal and fresh waters of China. Environ. Pollut. 221, 141–149.
- Karami, A., Golieskardi, A., Choo, C.K., Larat, V., Karbalaei, S., Salamatinia, B., 2018. Microplastic and mesoplastic contamination in canned sardines and sprats. Sci. Total Environ. 612, 1380–1386.
- Karami, A., Golieskardi, A., Choo, C.K., Larat, V., Galloway, T.S., Salamatinia, B., 2017. The presence of microplastics in commercial salts from different countries. Sci. Rep. 7.
- Karlsson, T.M., Vethaak, A.D., Almroth, B.C., Ariese, F., van Velzen, M., Hassellov, M., Leslie, H.A., 2017. Screening for microplastics in sediment, water, marine invertebrates and fish: method development and microplastic accumulation. Mar. Pollut. Bull. 122, 403–408.
- Katija, K., Choy, C.A., Sherlock, R.E., Sherman, A.D., Robison, B.H., 2017. From the surface to the seafloor: how giant larvaceans transport microplastics into the deep sea. Sci. Adv. 3.
- Kim, S.W., An, Y.J., 2019. Soil microplastics inhibit the movement of springtail species. Environ. Int. 126, 699–706.
- Kolandhasamy, P., Su, L., Li, J.N., Qu, X.Y., Jabeen, K., Shi, H.H., 2018. Adherence of microplastics to soft tissue of mussels: a novel way to uptake microplastics beyond ingestion. Sci. Total Environ. 610-611, 635–640.
- Kosuth, M., Mason, S.A., Wattenberg, E.V., 2018. Anthropogenic contamination of tap water, beer, and sea salt. PLoS One 13.
- Law, K.L., Thompson, R.C., 2014. Oceans. Microplastics in the seas. Science 345, 144–145.
- Lebreton, L.C.M., 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.
- Li, J.N., Qu, X.Y., Su, L., Zhang, W.W., Yang, D.Q., Kolandhasamy, P., Li, D.J., Shi, H.H., 2016. Microplastics in mussels along the coastal waters of China. Environ. Pollut. 214, 177–184.
- Li, Q.P., Sun, C.J., Wang, Y.J., Cai, H.W., Li, L.Y., Li, J.N., Shi, H.H., 2019. Fusion of microplastics into the mussel byssus. Environ. Pollut. 252, 420–426.
- Li, J.N., Yang, D.Q., Li, L., Jabeen, K., Shi, H.H., 2015. Microplastics in commercial bivalves from China. Environ. Pollut. 207, 190–195.
- Li, J.N., Green, C., Reynolds, A., Shi, H.H., Rotchell, J.M., 2018. Microplastics in mussels sampled from coastal waters and supermarkets in the United Kingdom. Environ. Pollut. 241, 35–44.
- Liebezeit, G., Liebezeit, E., 2013. Non-pollen particulates in honey and sugar. Food Addit. Contam. A. 30, 2136–2140.
- Liebezeit, G., Liebezeit, E., 2015. Origin of synthetic particles in honeys. Pol. J. Food Nutr. Sci. 65, 143–147.
- Liu, C.G., Li, J., Zhang, Y.L., Wang, L., Deng, J., Gao, Y., Yu, L., Zhang, J.J., Sun, H.W., 2019. Widespread distribution of PET and PC microplastics in dust in urban China and their estimated human exposure. Environ. Int. 128, 116–124.
- Lusher, A.L., Welden, N.A., Sobral, P., Cole, M., 2017. Sampling, isolating and identifying microplastics ingested by fish and invertebrates. Anal. Methods. 9, 1346–1360.
- Lusher, A.L., Burke, A., O'Connor, I., Officer, R., 2014. Microplastic pollution in the Northeast Atlantic Ocean: validated and opportunistic sampling. Mar. Pollut. Bull. 88, 325–333.

- Lusher, A.L., Tirelli, V., O'Connor, I., Officer, R., 2015a. Microplastics in Arctic polar waters: the first reported values of particles in surface and sub-surface samples. Sci. Rep. 5.
- Lusher, A.L., Hernandez-Milian, G., O'Brien, J., Berrow, S., O'Connor, I., Officer, R., 2015b. Microplastic and macroplastic ingestion by a deep diving, oceanic cetacean: the True's beaked whale *Mesoplodon mirus*. Environ. Pollut. 199, 185–191.
- Mason, S.A., Garneau, D., Sutton, R., Chu, Y., Ehmann, K., Barnes, J., Fink, P., Papazissimos, D., Rogers, D.L., 2016. Microplastic pollution is widely detected in US municipal wastewater treatment plant effluent. Environ. Pollut. 218, 1045–1054.
- Mathalon, A., Hill, P., 2014. Microplastic fibers in the intertidal ecosystem surrounding Halifax Harbor, Nova Scotia. Mar. Pollut. Bull. 81, 69–79.
- Muhlschlegel, P., Hauk, A., Walter, U., Sieber, R., 2017. Lack of evidence for microplastic contamination in honey. Food Addit. Contam. A 34, 1982–1989.
- Peller, J.R., Eberhardt, L., Clark, R., Nelson, C., Kostelnik, E., Iceman, C., 2019. Tracking the distribution of microfiber pollution in a southern Lake Michigan watershed through the analysis of water, sediment and air. Environ. Sci-Proc. Imp. 21, 1549–1559.
- Porter, A., Lyons, B.P., Galloway, T.S., Lewis, C., 2018. Role of marine snows in microplastic fate and bioavailability. Environ. Sci. Technol. 52, 7111–7119.
- Qiu, Q.X., Peng, J.P., Yu, X.B., Chen, F.C.Z., Wang, J.D., Dong, F.Q., 2015. Occurrence of microplastics in the coastal marine environment: first observation on sediment of China. Mar. Pollut. Bull. 98, 274–280.
- Rochman, C.M., Browne, M.A., Halpern, B.S., Hentschel, B.T., Hoh, E., Karapanagioti, H.K., Rios-Mendoza, L.M., Takada, H., Teh, S., Thompson, R.C., 2013. Classify plastic waste as hazardous. Nature 494, 169–171.
- Severini, M.D.F., Villagran, D.M., Buzzi, N.S., Sartor, G.C., 2019. Microplastics in oysters (*Crassostrea gigas*) and water at the Bahia blanca Estuary (Southwestern Atlantic): an emerging issue of global concern. Reg. Stud. Mar. Sci. 32, 12.
- Shen, L., Worrell, E., Patel, M.K., 2012. Comparing life cycle energy and GHG emissions of bio-based PET, recycled PET, PLA, and man-made cellulosics. Biofuel. Bioprod. Bior. 6, 625–639.
- Su, L., Deng, H., Li, B.W., Chen, Q.Q., Pettigrove, V., Wu, C.X., Shi, H.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.
- Taylor, R.B., Cole, R.G., 1994. Mobile epifauna on subtidal brown seaweeds in Northeastern New-Zealand. Mar. Ecol. Prog. Ser. 115, 271–282.
- Taylor, M.L., Gwinnett, C., Robinson, L.F., Woodall, L.C., 2016. Plastic microfibre ingestion by deep-sea organisms. Sci. Rep. 6.
- Thompson, R.C., Olsen, Y., Mitchell, R.P., Davis, A., Rowland, S.J., John, A.W.G., McGonigle, D., Russell, A.E., 2004. Lost at sea: Where is all the plastic? Science 304 838-838.
- Toussaint, B., Raffael, B., Angers-Loustau, A., Gilliland, D., Kestens, V., Petrillo, M., Rio-Echevarria, I.M., Van den Eede, G., 2019. Review of micro- and nanoplastic contamination in the food chain. Food Addit. Contam. A. 36, 639–673.
- Van Cauwenberghe, L., Vanreusel, A., Mees, J., Janssen, C.R., 2013. Microplastic pollution in deep-sea sediments. Environ. Pollut. 182, 495–499.
- Volkheimer, G., 1975. Hematogenous dissemination of ingested polyvinyl chloride particles. Ann. NY. Acad. Sci. 246, 164–171.
- Wesch, C., Bredimus, K., Paulus, M., Klein, R., 2016. Towards the suitable monitoring of ingestion of microplastics by marine biota: a review. Environ. Pollut. 218, 1200–1208
- Woodall, L.C., Sanchez-Vidal, A., Canals, M., Paterson, G.L.J., Coppock, R., Sleight, V., Calafat, A., Rogers, A.D., Narayanaswamy, B.E., Thompson, R.C., 2014. The deep sea is a major sink for microplastic debris. Roy. Soc. Open Sci. 1.
- Wright, S.L., Kelly, F.J., 2017. Plastic and Human Health: A Micro Issue? Environ. Sci. Technol. 51, 6634–6647.
- Yang, D.Q., Shi, H.H., Li, L., Li, J.N., Jabeen, K., Kolandhasamy, P., 2015. Microplastic Pollution in Table Salts from China. Environ. Sci. Technol. 49, 13622–13627.
- Yu, X.B., Peng, J.P., Wang, J.D., Wang, K., Bao, S.W., 2016. Occurrence of microplastics in the beach sand of the Chinese inner sea: the Bohai Sea. Environ. Pollut. 214, 722–730.
- Zhao, J.M., Ran, W., Teng, J., Liu, Y.L., Liu, H., Yin, X.N., Cao, R.W., Wang, Q., 2018. Microplastic pollution in sediments from the Bohai Sea and the Yellow Sea, China. Sci. Total Environ. 640, 637–645.