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The "Journey" of Microplastics across the Marine Food Web in China's Largest Fishing Ground

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Abstract: Microplastics in marine environments are becoming a hot topic since they can be transferred through the marine food web and may finally be consumed by humans. Here, we investigate the distribution characteristics of microplastics in marine organisms at different trophic levels through their digestive tracts (entire organisms for zooplankton and zoobenthos). A total of 124 fish and 22 crustaceans from 10 fish and 3 crustacean species, as well as a few zooplankton and zoobenthos, were captured from the Zhoushan fishing ground, i.e., China's largest ocean fishing ground. The abundance of microplastics ranged from 0.74 ± 1.29 to 4.71 ± 2.19 items per sample in fish species and from 0.83 ± 1.07 to 1.00 ± 0.93 items per sample in crustacean species. Among the detected microplastics, fiber was the most dominant type (i.e., 67%), transparent microplastics were the most frequently detected (i.e., 49%), and the majority of the microplastics were identified as natural particles (cellulose). The abundance of microplastics was positively correlated with the trophic level (correlation coefficient = 0.717; $p < 0.05$). Our results show that microplastics are widespread in the marine organisms of the Zhoushan fishing ground, and they might accumulate in marine organisms at higher trophic levels of the marine food chain.

Keywords: microplastics; Zhoushan fishing ground; marine organism; food chain; natural microplastics

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1. Introduction

Plastic pollution is a growing global concern and constitutes a crucial threat to the marine environment [1–3]. Generally, the integrity of plastics will decrease via physical, chemical, and biological interactions in the ocean [4], leading to the formation of plastic particles ranging in size from meters to nanometers. Previous studies have revealed that microplastics (defined as plastic pellets with a diameter of <5 mm) are widespread in marine environments and have accumulated across a range of habitats [5–7]. Moreover, during the past few years, research on microplastics has gained substantial momentum. They have been detected in the Southern Ocean [8], Arctic sea ice [9], the Pacific Ocean [10], the East China Sea [11], the Atlantic Ocean and the Indian Ocean [12], and marine ranching areas [13]. Peng et al. [14] found that microplastic abundance in hadal sediments of the Mariana Trench varies from hundred to thousand pieces/L, which is extraordinarily higher than that in other deep-sea sediments, suggesting that plastics have contaminated even the deepest and most remote places on Earth. Desforges et al. [15] reported that microplastic concentrations ranged several orders of particles/m^3 in subsurface seawater in the northeast Pacific Ocean. Notably, in estuaries and offshore areas, the concentration of microplastics is relatively higher than that in the open sea [11].

Because of their small size, microplastic uptake has been identified in a variety of marine taxa, including plankton, bivalves, polychaetes, crustaceans, fish, turtles, and seabirds [16–20], thereby covering almost all trophic levels and feeding modes, from planktonic organisms [18] to top predators [21]. The ingestion of microplastics by marine organisms may lead to adverse physical and physiological effects. Studies have reported that the uptake of microplastics by birds, fish, as well as invertebrates attenuates their foraging behavior and feeding habitats, thereby leading to trophic barriers between them [22–24]; notwithstanding, these studies used much higher particle concentrations than those naturally occurring in the environment. Moreover, microplastics will inevitably suffer physicochemical and biological aging processes [25], which can facilitate the release of additives mixed with monomers into the environment and increase the adsorption of harmful ingredients from their surroundings, such as toxic metal ions and persistent organic pollutants (POPs) [26–28]. Another issue worthy of mention is that pathogens or invasive microbes can colonize microplastics, resulting in their spread with the movement of microplastics [29]. All of the above are attributed to the potential toxicity of microplastics and may lead to severe ecological risks [17].

According to artificial food chains [18,30,31], the trophic transfer of microplastics was identified and quantified through in-field organisms, their natural predators, and controlled feeding experiments. Initially, small plastic particles were identified in the scats of *Phocarctos hookeri* in conjunction with otoliths of *Electrona subaspera* [32]. The pioneering study on the trophic transfer of microplastics was a study of the discovery of microplastics in sea lion scats a decade ago [33]; it revealed that human-made polystyrene nanoparticles are transported across an aquatic food web, starting from algae to zooplankton and finnaly to fish, thereby affecting the behavior of the top consumer. It was suggested that plastic concentrations in the water environment significantly underestimate exposure levels, which is an important consideration for future risk assessment studies [34].

In this study, we investigate the abundance and distribution of microplastics across different species and identify the association between microplastic abundance in marine animals belonging to different trophic levels sampled from the Zhoushan fishing ground. This study will contribute to the complete microplastic distribution database regarding marine organisms in China.

2. Materials and Methods

2.1. Sample Collection

All samples were collected using a bottom trawl from July to October 2019 at 26 sampling stations across the Zhoushan fishing ground (Figure 1). Bottom trawl nets (32 m long; 22 mm mesh size) were used to conduct trawl surveys in the study area according to the "Marine Survey Specification" (GB/T 12763.6-2007). The average towing speed of fishing boats during sampling was maintained at 3.1 knots/h. A total of 146 marine organisms were captured, including 124 individuals from 10 fish species, 22 individuals from 3 crustacean species, and a few zooplankton (belonging to Copepoda) and zoobenthos (belonging to Nereidae). For each species, habitat and trophic levels were classified according to the FishBase database [35]. The names and classifications of each marine organism were recorded according to the China Marine Biology Directory and the SealifeBase database, and the ecological behaviors of these marine organisms (Table 1) were fully investigated and recorded. Before the extraction of microplastics, all organisms were weighed using an electronic balance and measured using a Vernier caliper. The digestive tracts of the fish and crustacean samples were dissected, while intact individuals of Copepoda and Nereidae were preserved. Different samples were placed in different glass bottles and stored in a refrigerator at −20 °C until further analysis.

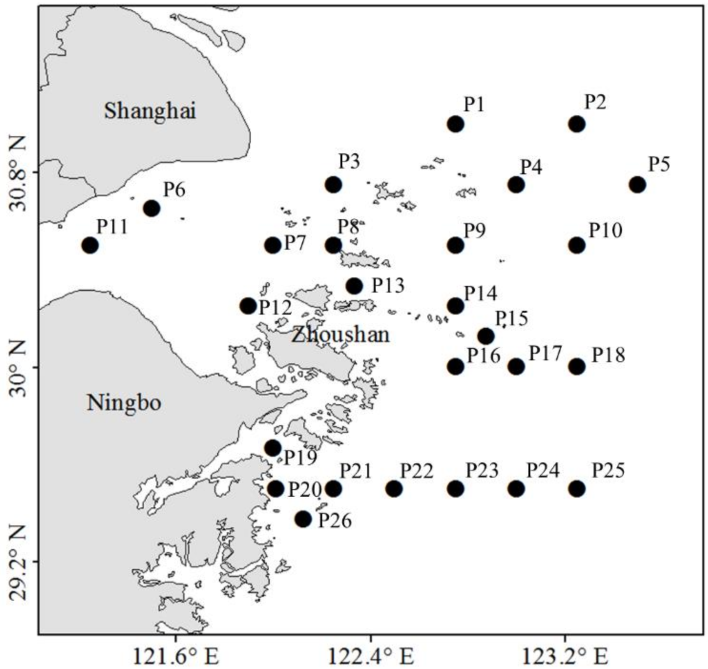

Figure 1. Sampling locations at the Zhoushan fishing ground between July and October 2019.

Table 1. Basic biological data and trophic levels of captured marine organisms in the Zhoushan fishing ground.

	Species	Feeding Features	Total Samples	Body Weight ± SD (g)	Fork Length ± SD (mm)	Trophic Level
Fish	*Harpodon nehereus*	Benthos; nekton	18	68.92 ± 42.07	210.17 ± 40.37	3.81
	Collichthys lucidus	Benthos; nekton	17	21.92 ± 7.84	117.83 ± 14.83	3.29
	Scomber japonicus	Plankton; nekton	8	179.72 ± 89.13	239.79 ± 48.77	3.30
	Pampus argenteus	Plankton; jellyfish	12	96.13 ± 61.34	148.29 ± 33.97	3.10
	Larimichthys polyactis	Benthos; nekton	10	40.02 ± 20.50	136.00 ± 18.38	3.46
	Trichiurus lepturus	Nekton; crustacean	7	143.31 ± 119.09	186.60 ± 57.12	3.82
	Cynoglossus trigrammus	Benthos	14	106.16 ± 13.75	251.60 ± 2.87	— [a]
	Sardinella zunasi	Plankton	5	5.36 ± 1.19	75.13 ± 3.72	2.66
	Muraenesox cinereus	Benthos; nekton	6	182.41 ± 123.79	200.00 ± 47.34	3.85
	Johnius belengerii	Benthos; nekton	27	71.95 ± 40.37	149.83 ± 28.26	3.53
Crustacean	*Charybdis japonica*	Benthos	9	72.70 ± 30.99	51.33 ± 7.94	2.77
	Portunus trituberculatus	Benthos	6	68.28 ± 71.58	50.17 ± 15.25	2.85
	Exopalaemon carinicauda	Benthos	7	5.06 ± 0.72	75.14 ± 3.09	3.16
Copepoda [b]	—	—	0.2 g	—	—	2.17
Nereidae [b]	—	—	0.2 g	—	—	2.22

Notes: [a] The trophic level was not investigated in any previous study; [b] Copepoda and Nereidae has not been further classified.

2.2. Digestion Procedures

Microplastics were extracted from marine organisms according to the method of Masura et al. [36], with some modifications. The digestive tracts of fish and Crustacea and 0.2 g each of zooplankton and zoobenthos samples were transferred into 250 mL pre-cleaned glass beakers separately, and the glass beakers containing the samples were dried at 60 °C in a drying oven for 12 h. To avoid the influence of organic matter, 20 mL of ferrous sulfate solution (0.05 M) (AR, Aladdin, China) was added to the sample in each glass beaker, followed by the addition of 20 mL of 30% H_2O_2 (Sinopharm, China). The glass beakers were placed at 60 °C in a thermostatic water bath; the solutions were stirred simultaneously and then incubated for 4 h at 60 °C to complete the reaction. NaCl was added to the solution at a 3 g/10 mL concentration until it reached saturation (~5 M NaCl); the solution was then manually stirred with a clean glass rod for 2 min. The mixture was kept undisturbed for 4 h at 60 °C. The clean supernatant was transferred onto a piece of 8 µm pore-size, 47 mm diameter glass-fiber filter paper (Shanghai Xingya, China) with the assistance of a vacuum pump (SHB-III, Kewei Yongxing, Beijing, China). All filter instruments were rinsed with Milli-Q water several times, and the washing solutions were filtered through the same glass filter. Finally, the filter was placed on a clean petri dish and was air-dried at room temperature for microscopic inspection.

2.3. Microscopic Inspection

The microplastics were thoroughly determined by adopting a z-shaped pattern from left to right with a digital microscope (Sunny Optical Technology, China) equipped with MvImage software, with up to ×160 magnification, aiming to classify the microplastics based on their external characteristics. The particles were visually identified, and the color, shape, and size of the suspected microplastics were recorded for further analysis (Figure S1). The shapes of the particles were separated into four categories: fiber, fragment, film, and foam. Particle color was classified into five groups: transparent, black, blue, red, white, and yellow. Size >100 µm was selected for the µ-FTIR analysis according to operational experience during the experiment.

2.4. Identification and Validation of Microplastics with µ-FTIR

A total of 91 representative plastic-like particles on the filter paper were selected based on their appearance (e.g., colors, shapes, and sizes) with forceps for validation using a micro-Fourier transform interferometer (µ-FTIR; Nicolet™ iN10; Thermo Scientific, Waltham, MA, USA) in reflection mode. The detector was cooled with liquid nitrogen prior to use. Microplastic particles were selected using a needle and were subsequently placed on the surface of the gold plating slide. The detector was operated in the 675–4000 cm^{-1} wave number range, with a 3 s collection time and the co-addition of 16 scans at 8 cm^{-1} resolutions. The aperture was set to 16–20 µm. The spectra were obtained using OMNIC™ Picta™ software (Thermo Fisher Scientific) and compared with the OMNIC polymer spectra library to verify the polymer type (Table 1). The spectrum analysis followed the method of Woodall et al. [37], where a matching quality index of ≥70% with a standard database was acceptable.

2.5. Quality Controls of Experiment

Experimental apparatuses were rinsed with Milli-Q water three times and then dried in a drying oven before the experiment. The plastic container was replaced with a non-plastic container, while the laboratory window remained closed during the experiment to avoid airflow and the experimental process was implemented in a clean fume hood (F8P150-001, Scienfocus, Guangzhou, China). Procedural blanks, aiming to detect environmental microplastic contamination, were carried out with the same isolation and digestion procedures used for animals but using Milli-Q water and glass microbeads of 1 mm diameter instead of digestive tracts. In summary, 0.008 ± 0.014 items mL^{-1} with fiber shapes were detected in the triplicate Milli-Q water blanks, indicating low contamination from lab airborne sources. The recovery rates were assessed as previously described [13]. Briefly, commercial plastic beads were selected as the reference particles and mixed with glass microbeads, and then the mixture was applied to the floating separation process. The recovery rate of microplastic particles was about 96%.

3. Results

3.1. Abundance and Distribution of Microplastics in Marine Organisms

Among the 124 fish, 22 crustaceans, and several small zooplankton and zoobenthic organisms, microplastics were detected in 96 individuals based on μ-FTIR analysis, accounting for 66.2% of the total number of samples, and 232 particles were identified as polymers. In total, 210 microplastic particles were detected in fish, and the abundance of microplastics ranged from 0.74 ± 1.29 to 4.71 ± 2.19 items/digestive tract (Figure 2). The highest abundance of microplastics was 8 items/digestive tract, detected in *Muraenesox cinereus*. Furthermore, 20 microplastic particles were detected in crustaceans, and the abundance of microplastics ranged from 0.83 ± 1.07 to 1.00 ± 0.93 items/digestive tract (Figure 2). The highest abundance of microplastics was 3 items/digestive tract, detected in *Portunus trituberculatus*. Only two microplastic particles were detected in zooplankton and zoobenthic organisms, with a 0.50 ± 0.50 per 0.1 g^{-1} average abundance.

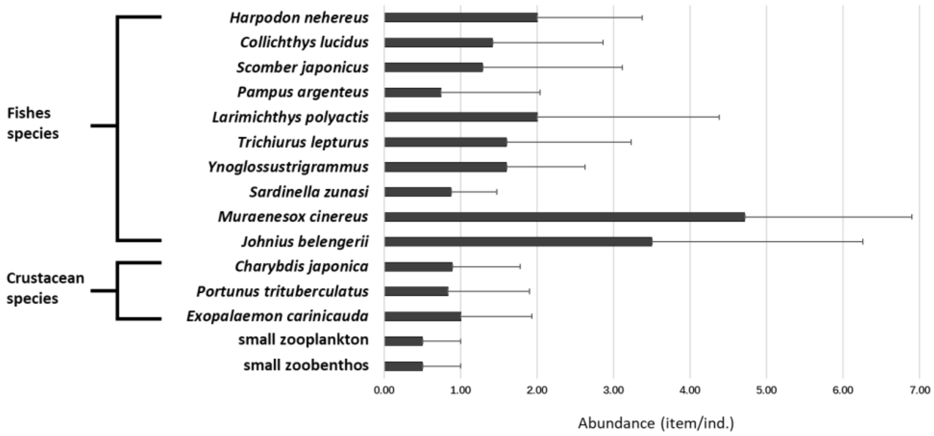

Figure 2. Average abundance of microplastics (±SD) in the gastrointestinal tracts of fish and crustaceans and the body of zooplankton and zoobenthos from the Zhoushan fishing ground.

3.2. Physicochemical Characteristics of Microplastics

The most common microplastic shape was fiber (67%), followed by film (19%), fragment (13%), and foam (1%) (Figure 3a). Regarding microplastic color, transparent (49%) particles were the most dominant, followed by black (25%), red (12%), and blue (11%) (Figure 3b). Green and white microplastics were less observed.

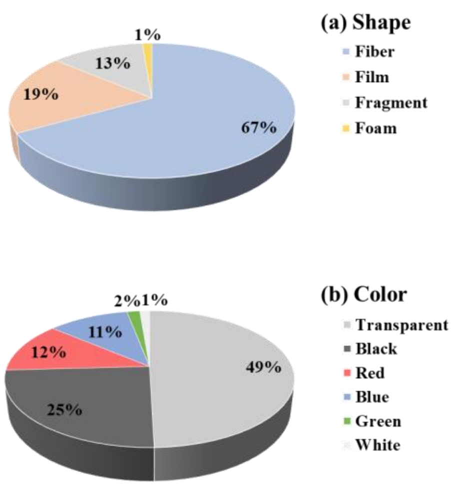

Figure 3. Microplastic abundance in marine organisms of the Zhoushan fishing ground, categorized by (**a**) shape and (**b**) color.

In total, 232 particles were determined as polymers using μ-FTIR spectroscopy (Table 2). Natural microplastics (i.e., cellulose) were the dominant type, accounting for 50.4% of the total elements, followed by synthetic polymers: cellophane (22.4%), rayon (9.9%), polyethylene terephthalate (PET) (6.9%), polyester (3.9%), acrylic (3.0%), polystyrene (PS) (1.3%), polyethylene (PE) (0.9%), polypropylene (PP) (0.9%), and polyamide (PA) (0.4%).

Table 2. Information on the identified microplastics.

Type of Polymers	**Number**	**Proportion of Total Particles (%)**	**FTIR Library**
Cellulose	117	50.4	Wizard Library
Cellophane	52	22.4	Hummel Polymer Sample Library
Rayon	23	9.9	Synthetic Fibers by Microscope
Polyethylene terephthalate	16	6.9	Cross Sections Wizard
Polyester	9	3.9	Synthetic Fibers by Microscope
Acrylic	7	3.0	Synthetic Fibers by Microscope
Polystyrene	3	1.3	Aldrich Linked IR
Polyethylene	2	0.9	Hummel Polymer Sample Library
Polypropylene	2	0.9	Polymer Additives and Plasticizers
Polyamide	1	0.4	Hummel Polymer Sample Library
Total	232	-	-

3.3. Transfer of Microplastics across Marine Food Webs

The trophic level of each species was estimated through a comprehensive analysis of the food-web structure of the Zhoushan fishing ground. The trophic level of each species was taken as the average of those assigned by previous studies [38–45] and is listed in Table 1. For example, the trophic level of *Collichthys lucidus* was 2.93, 3.282, 3.5, 3.03, and 3.7, respectively, in the above studies, and thus, the corresponding average value (TL = 3.29) was used in this study.

According to Yu et al. [44], *Hapodon nehereus* (Bombay duck) is the dominant species in the Zhoushan fishing ground throughout the year. Meanwhile, *Exopalaemon carinicauda* (ridgetail white prawn) and *P. trituberculatus* (swimming crab) are economically important crustaceans. They are not only the dominant species in offshore fisheries but also two of the most important fishery resources in the coastal areas of China. Thus, two typical feeding relationships were selected as follows: 1) *M. cinereus* (conger pike)—*Trichiurus lepturus* (cutlassfish)—*P. trituberculatus* (swimming crab), small zoobenthos; 2) *H. nehereus* (Bombay duck)—*Collichthys lucidus* (spinyhead croaker)—*E. carinicauda* (ridgetail white prawn)—small zooplankton. *M. cinereus*, *T. lepturus*, *H. nehereus*, and *C. lucidus* were the higher trophic predators, whereas *P. trituberculatus*, *E. carinicauda*, small zoobenthic organisms, and zooplankton were the prey. *M. cinereus* had the highest trophic level, corresponding to the highest average abundance of microplastics detected; small zooplankton and zoobenthic organisms had the lowest trophic level, corresponding to the lowest average abundance of microplastics detected.

The relationship between trophic level and microplastic abundance (quantified as items/digestive tract) is shown in Figure 4. A high correlation between microplastic abundance and trophic level was identified (Spearman correlation coefficient: 0.717; $p < 0.05$), with microplastic abundance increasing with increasing trophic level (Figure 4). Microplastic abundance in crustacean species was significantly lower than that in fish species ($p < 0.05$), and the trophic levels of crustacean species were obviously lower than those of fishes (Figure 4).

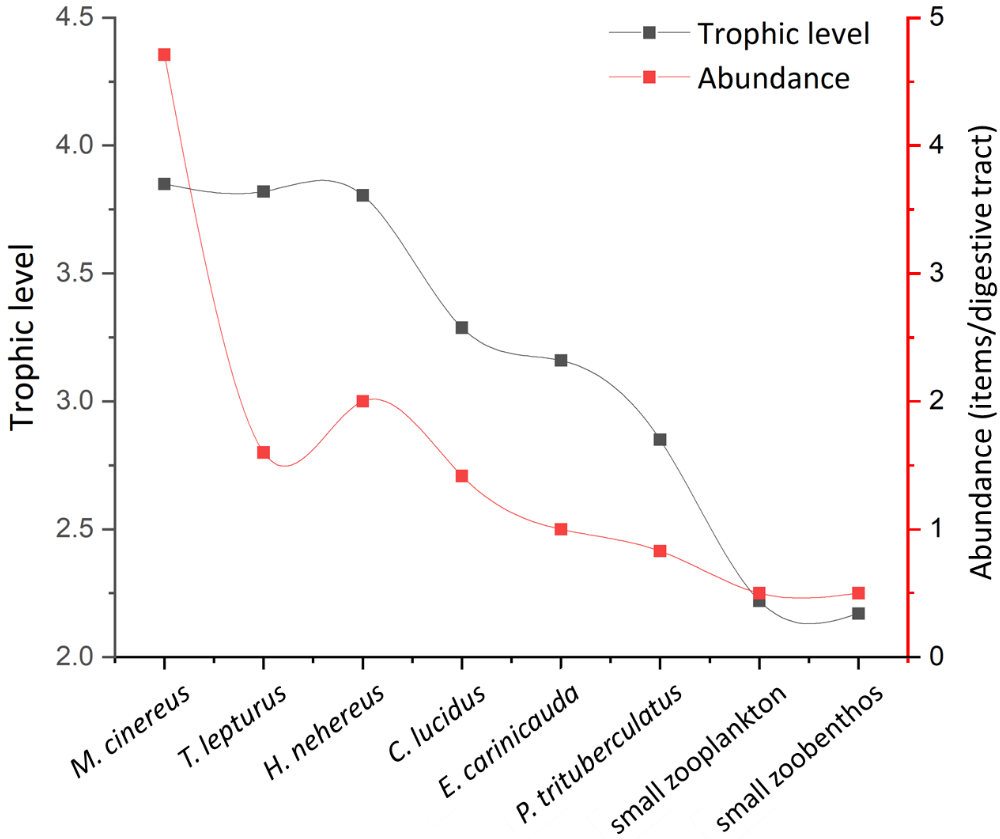

Figure 4. Relationship between trophic level and microplastic abundance (quantified as items/digestive tract) (correlation coefficient: 0.717; $p < 0.05$).

4. Discussion

4.1. Abundance of Microplastics in Different Species

We investigated the abundance of microplastics accumulated by 13 major marine species from the Zhoushan fishing ground and detected microplastics in 66.2% of the digestive tract samples. All species were found to have ingested microplastics, which might be connected with the wide distribution of microplastics in the surrounding marine environment. For instance, the mean microplastic concentration was 0.18 ± 0.05 (items/g sediment) in Hangzhou Bay [46] and 144 items/m^3 in the coastal surface seawater of Zhejiang Province [47]. Our results suggested that marine fish may be important intermediate carriers in the marine environment. Moreover, the abundance of microplastics in the digestive tract of fish was also compared with the results reported worldwide (Table 3). The average abundance of microplastics found in the digestive tracts of fish was 1.69 ± 1.97 items/digestive tract, which was about three times higher than that found in fish captured in the East China Sea [48]; this variation may be due to the different statistical types of microplastics. For example, the two most abundant microplastics were cellulose and cellophane in the present study, both of which were not calculated by Zhang et al. [48]. It should be noted that some related studies excluded cellulose [49], whereas others did the opposite [50–52]. In the present study, cellulose is also considered due to its high abundance. The cellulose was also predominant (60–88% of microplastics) in all commercial species in Xiangshan Bay (geographically near the Zhoushan fishing ground), including fish, bivalves, and shrimps [53], suggesting that its contamination in marine animals should not be neglected. In any case, the results of this study provide fundamental data for future toxicology research, which can help to reveal the toxic effects of marine microplastics on marine animals.

Table 3. Concentration of microplastics (MPs) ingested by fish in the natural environment.

Region	Number of Species	MP Concentration (MPs/Fish)	Reference
Europe	26	0.27 ± 0.63	[54]
UK	10	1–15	[55]
North Pacific	6	1–83	[56]
Xiangshan Bay	2	1.8–2.1	[53]
East China Sea	11	0.52 ± 0.35	[48]
Zhoushan Fishing Ground	10	1.69 ± 1.97	This study

4.2. Morphotype, Color, and Chemical Composition of Microplastics

Fibrous microplastics accounted for the highest proportion (67%) of morphotypes in this study. Fibers are formed mainly by the degradation of larger plastics under the combined action of oxygen, ultraviolet light, heat, and mechanical effects such as wind. In fact, a large number of studies have shown that fibrous microplastics are the main source of microplastic pollution [17,53,57–59], which affects even deep-sea sediments and organisms [10]. Fibers in wastewater from sewage treatment plants in urban areas may be associated with certain household activities, such as washing-machine wastewater discharge through sewage treatment plants releasing large amounts of microplastics into the coastal region [60,61]. Among the colors of microplastics observed in this study, transparent microplastics were the most abundant, accounting for 49% of the total particles, followed by black (25%), red (12%), blue (11%), green (2%), and white (1%) (Figure 3b). Color plastic particles with a spectrum of darkening tones are considered to carry more organic pollutants [62], and they can attract predators because they are similar in color to their prey. The blue microplastics were probably derived from the employment of plastic fishing gear around the Zhoushan fishing ground. Regarding the chemical composition of microplastics, 10 types of polymers were identified: cellulose, cellophane, rayon, PET, polyester, acrylic, PS, PE, PP, and PA. These plastics generally originate from a variety of household and industrial applications. Among all detected microplastic particles, cellulose and cellophane were the most frequently found. Cellulose is a natural polymer; it is thought to be derived from anthropogenic sources (e.g., textile waste) [63]. Suaria et al. [64] emphasized the comparatively high numbers of natural and cellulosic fibers (91.8%) present within water columns around the world. Cellophane is an organic cellulose polymer that is widely used for food packaging and as a release agent for glass-fiber rubber [65], and it was found to be the main microplastic type at several sampling sites (such as in the estuary of the United Kingdom) [66]. Studies have shown that cellophane accounts for the highest proportion of microplastics in sediment and fish samples from the coast of the East Sea and the Ma'an Archipelago [11,13]. In marine ecosystems, fishery products, such as fishing lines and nets, may be the sources of PA, PE, and PP.

4.3. Microplastic Transfer across Trophic Levels in the Marine Food Chain

The highest concentration of microplastics was found in the digestive tracts of *M. cinereus* and *Johnius belengerii* (Figure 2), indicating that these two species ingest microplastics more frequently than the other marine organisms or that microplastics can gradually accumulate in marine organisms at higher trophic levels through the marine food web. *M. cinereus* (TL = 3.85), *T. lepturus* (TL = 3.82), and *H. nehereus* (TL = 3.81) took up the highest trophic niche in this study, followed by *J. belengerii* (TL = 3.53), *Larimichthys polyactis* (TL = 3.46), *Scomber japonicus* (TL = 3.30), *Collichthys lucidus* (TL = 3.29), *E. carinicauda*, *Pampus argenteus*, *P. trituberculatus*, *Charybdis japonica*, and small zoobenthos and zooplankton (Table 1). The amount of microplastics extracted from organisms at each trophic level generally increased with an increase in trophic level. This study is consistent with [48], which concluded that the trophic level may be the transport pathway for microplastics from low to high trophic-level predators. In addition, microplastics were detected in the digestion of zooplankton and zoobenthos. Zooplankton consists of economically important aquatic organisms and constitutes an important food source for marine organisms in the upper and middle waters; microplastics are likely to enter the bodies of organisms with higher trophic levels through the food chain. Setälä et al. [18] found that plastic particles can be ingested by different plankton groups, and microplastics may have multiple transport pathways in the upper mesoporous food webs, suggesting that high concentrations of microplastic pollution may accumulate in marine food webs. Microplastics can be identified in various parts of the marine food web through multiple pathways.

Generally, the biomagnification of microplastics in the marine food web can hardly be concluded based on our results. Previous studies have found that concentrations of microplastics decrease as the trophic level increases [67,68], based on a bioaccumulation model using a mass balance of uptake and loss rates. Wang et al. [69] found that there was no biomagnification of microplastics by comparing crabs with mussels, and they speculated that the egestion of ingested microplastics by marine animals might affect related results. Likewise, the slight increase in particles with trophic level in the present study could also be explained by increases in food requirements depending on body size. Moreover, because microplastics can be concentrated in certain organs and tissues, analyzing only the abundance of microplastics in specific tissues may lead to incomplete or inaccurate conclusions. Even so, the present result clearly suggests that microplastic abundance will increase in the digestive tract of marine animals as the trophic level increases. Currently, the information on the relationship between microplastic abundance and trophic level is limited, and whether there is a positive correlation between the amount of ingested or accumulated microplastics and the trophic level requires further research.

5. Conclusions

Microplastic pollution in marine environments affects various organisms in the ocean. The entry of microplastics into the marine food chain has led to the accumulation of microplastics in top predators. The results of this study reveal that microplastics are present in the digestive tracts of fish and crustaceans, as well as in small zooplankton and zoobenthos. The chemical composition, morphotype, and size of the detected microplastics were different. However, it is unclear whether microplastics in the benthic or plankton food chain are biologically amplified or diluted at different trophic levels. In the future, experiments could be conducted with specific sample sizes and more realistic concentrations of microplastics on a lab scale to draw more credible conclusions. In any case, the results of this study provide fundamental data for future toxicology research, which can help to reveal the toxic effects of marine microplastics on marine animals. Furthermore, highlighting effective treatment measures for alleviating marine plastic pollution is also urgent.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/w15030445/s1. Figure S1: Some microscopic inspection images of microplastics. (a) Blue fiber; (b) transparent film; (c) black fragment.

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Conflicts of Interest: The authors declare no conflict of interest.

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