



Research Article

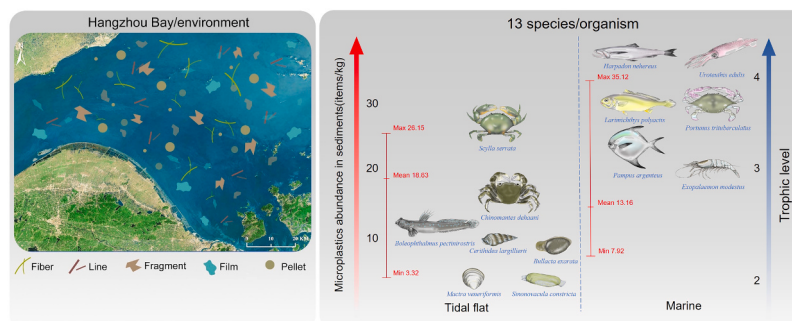
Coupling of microplastic contamination in organisms and the environment: Evidence from the tidal flat ecosystem of Hangzhou Bay, China

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HIGHLIGHTS

- The study revealed high levels of microplastic pollution in Hangzhou Bay, China.
- Different types of microplastics were found, including fibers and fragments.
- Spatial distribution patterns of microplastic abundance were showed in Hangzhou Bay.
- A significant correlation was found between microplastics in organisms and sediments.
- The results emphasize the need for strategies to reduce microplastic pollution.

GRAPHICAL ABSTRACT



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ABSTRACT

Microplastics are a new type of contaminant, widely defined as fragmented plastics with the longest dimension or diameter less than 5 mm, that are widely distributed, difficult to degrade, and easily adsorb other pollutants. Estuaries are key habitats where terrestrial microplastics flow in water runoff and import into the ocean. The ubiquitous use of plastics has resulted in a massive amount of plastic waste that is released and accumulated in bay ecosystems, posing serious ecological impacts. The study of microplastic contamination in Hangzhou Bay, the estuary of the Qiantang River, has important theoretical value in ecology and environmental science. Microplastic contamination in the tidal flats and organisms of Hangzhou Bay is serious and microplastic characteristics (type, size, and polymer type) in organisms were significantly correlated with those in the environmental media. Spatial autocorrelation was found in the abundance of microplastics in marine and tidal flat sediments of Hangzhou Bay, China, but no spatial autocorrelation was found in the sediment environment as a whole. The microplastic abundance in each organism in this study was not statistically correlated by weight or by individual count with its corresponding trophic level ($P = 0.239 > 0.05$; $P = 0.492 > 0.05$, respectively). Our study suggests a coupling relationship of microplastic contamination between organisms and the environment

Abbreviations: PP, Polypropylene; ABS, Acrylonitrile-Butadiene-Styrene; AA, Acrylic Acid; CP, Cellophane; CA, Cellulose Acetate; LDPE, Low-Density Polyethylene; HDPE, High-Density Polyethylene; PS, Polystyrene; PP+PE, Polypropylene+Polyethylene; PPC, Polypropylene Copolymer; PAA, Poly(Acrylic acid); PET, Polyethylene Terephthalate; PVT, Polyvinyltoluene; PMP, Polymethylpentene; PVC, Polyvinyl Chloride; PU, Polyetherurethane; PA, Polyamide; PI, Polyimide; PE, Polyethylene; PL, Polyester; HEC, Hydroxyethyl Cellulose; RY, Rayon; CE, Cellulose; μ -FTIR, μ -Fourier Transform Infrared Spectroscopy.

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and can provide essential data and a scientific foundation for the study of microplastics pollution in Hangzhou Bay, as well as provide important evidence for the ecological and health risk assessment of microplastics.

1. Introduction

Marine plastic litter has become an ecological pollution problem of global concern due to the large and increasing threat it poses to marine and coastal tidal flat ecosystems. After being exposed to long-term physical, chemical, and biological processes in the marine environment, plastic litter is eventually broken down into plastics with maximum diameters less than 5 mm, known as “microplastics”. The fragmentation of large, individual plastic items leads to the creation of millions of microplastics, which are thought to be the direct source of most of plastic debris in the ocean today [1]. The emergence of microplastics in the environment may pose an ecological threat to human existence.

More than 80% of marine plastic waste comes from land [2]. The developed economies of coastal areas, along with the large populations close to the coastline, generate land-based sources of plastic waste that enter the marine ecosystem directly or indirectly through sewage systems, rivers, the atmosphere, and human activities (tourism and aquaculture) [3]. Estuaries are key areas where terrestrial microplastics flow with water runoff and import into the ocean [4,5]. The process of flow diffusion in estuarine habitats is complex, and is further complicated by the influence of tides, waves, coastal currents, estuarine topography, and evolution. Therefore, the special geographical environment of urban estuarine habitats increases the possibility of microplastic deposition and accumulation, leading to high microplastic load in estuarine habitats. The high load of microplastics in estuarine habitats is not only manifested in environmental media, but also in the organisms that inhabit estuarine ecosystems, such as the sandworms of Haeju Bay and its intertidal zone in North Korea, the Xiangshan Harbor and its farmed fish in China, the Cienfuegos Bay and its local *Perna viridis* in Cuba, and the Terengganu estuary and its zooplankton in Malaysia [6–9]. Typical indicator organisms, such as sand worms, also actively ingest microplastics and the degree of microplastic pollution in their bodies can reflect the degree of microplastic pollution in habitats overall, such as tidal flats and estuaries where microplastic pollution is high [9].

Gulf estuaries are highly productive ecosystems with a large amount of nutrient turnover. Upstream deposition of man-made products transport large amounts of organic debris, which usually accumulate in coastal and off-shore fisheries around the mouth of bays, such as Newfoundland fishery in Canada and Zhoushan fishery in China. These ecosystems bring together a great diversity of aquatic species that are freshwater, marine, and those that migrate between both bodies of water. The aquatic products of these systems (e.g., shellfish, crabs, shrimp, fish, seaweed) are important sources of food for humans. Bay estuary areas are not only an economically developed areas, but also ecologically vulnerable zones that are crucial to sustainable development [10]. Identifying microplastic contamination in aquatic life in coastal regions is particularly important to analyze the threat of microplastics to human health. Understanding how microplastics affect estuarine life is therefore of interest not only to biologists, but increasingly to policy makers and environmental managers.

Most studies across the globe have reported the presence of microplastics in freshwater, coastal environments, and organisms [11–16], but few studies have focused on the relationship between marine sediments, organisms, and microplastic contamination in the surrounding tidal flats. Similarly, few studies have attempted to track the fate of microplastics and mixed contaminants through the complex marine food web using environmentally relevant concentrations to identify the true level of risk [17]. One study indicated that microplastic pollution in cultured marine economic species in Xiangshan Bay of China was related to trophic level changes, but microplastic impacts on commercial species

may be less harmful than previously thought [7]. A correlation between microplastic community composition in different habitats was also observed to gradually weaken with the increase in point source distance [18]. Several experimental laboratory-based studies have been conducted but do not fully reflect the true field ecosystem and may have limited applicability to the natural environment, due to crucial differences between the characteristics of microplastics in fish in a laboratory environment and the natural environment [17,19]. However, there are studies that suggest that the trophic level of fish is one factor that influences microplastic uptake in fish [20]. In contrast, another study found no microplastic accumulation in the food web [21]. Microplastics in mangrove habitats appear to transfer between different trophic levels and are bioamplified in successive trophic levels [22]. However, a different study suggested that the microplastic pollution in benthic organisms is not affected by the changes of biological habitat or trophic level [23]. Therefore, by collecting wild aquatic organisms from natural habitats of estuaries and testing whether the microplastic content in them is related to the microplastic content deposited in the environment of the same site based on their trophic levels, the sources and entry routes of microplastics in estuaries can be identified. In turn, these results may inform the local protection of marine fisheries.

Hangzhou Bay is an offshore tidal flat ecosystem in China. It is an estuary of Qiantang River that is also affected by runoff from the Yangtze River. Here, sediment and organic particles carried by the rivers interact with ocean currents of the Pacific Ocean to form one of the world's largest fisheries — the Zhoushan fishery. The aims of this study are: 1) to examine the microplastic pollution in organisms in the estuary ecosystem of Hangzhou Bay and analyze its spatial correlation with environmental microplastic pollution; 2) to explore the relationship between microplastic pollution and trophic level of marine economic species of Hangzhou Bay and whether there is a biomagnification of microplastics; 3) to quantify the load of microplastics in marine species of commercial interest located in the coastal ecosystem of Hangzhou Bay and assess the risk of microplastic accumulation in humans through consumption of marine food products.

2. Study area and methods

2.1. Overview of the study area

Hangzhou Bay, located in the northeastern part of China's Zhejiang province, has the largest tidal difference of any bay along the coast of China, and is one of the eight largest estuaries in China. It contains a typical coastal wetland ecosystem with a tidal flat area of about 550 km² and is listed as an important wetland in China. Hangzhou Bay is rich in nutrients, fertile water, bait organisms, and hydrological environment, and has many large fishing grounds, rich fishery products, and a developed fishery economy. Hangzhou Bay is also an important stepping-stone along the migration route of East Asian migratory birds, and a large number of wetland water birds inhabit this location [24].

In recent decades, China's economic and social development has put tremendous pressure on the coastal waters around Hangzhou Bay due to pollution. As early as 2003, the Zhejiang provincial government included the six cities along the coast into the Hangzhou Bay area, which now account for 79.80% of Zhejiang's total economic output. Hangzhou Bay is located in a highly urbanized area of eastern China that includes a variety of industries (e.g., chemical, dyeing, and food factories) and hydroelectric power plants [24]. The Hangzhou Bay region has experienced increased urbanization, industrialization, and agricultural development. These human and economic activities have resulted in the import of large amounts of harmful pollutants into Hangzhou Bay [25],

leading to the continued deterioration of environmental quality in the region [26].

2.2. Sample collection

Employing a grid sampling method, a grab sampler (Van Veen, Hydro-Bios, Germany) was used to collect 5 cm of surface depth marine sediment at stations M 1–13 in the summer of July 2020 (see Fig. 1). Tidal flat sediment samples at 5 cm surface depth were collected at stations T 1–5 in the summer of July 2021 using a shovel, and the samples were stored in an aluminum box (see Fig. 1).

Several organism samples were caught directly in the tidal flats of Hangzhou Bay in July 2021 and several samples of organisms from Hangzhou Bay were caught by local fishermen in October 2021. The legends and specific information about the organism samples are shown in Annex Table S1 and Fig. S1.

2.3. Extraction and identification of microplastics

Three sediment parallel samples were taken from each site and the sediment samples (about 160 g) were placed in a 60 °C oven to dry to constant weight. Saturated 300 mL NaCl solution (at least 400 g of NaCl was added into 1 L Milli-Q water at 40 °C. The solution was stirred until the NaCl was precipitated. After cooling at room temperature, the NaCl crystals were resolved to determine the saturated NaCl solution) was added for flotation and stirred well to cause the plastic sample float on the liquid surface. After standing for at least 2 h, supernatant was transferred to another beaker and the above steps were repeated 3 times for each sample to improve the plastic recovery rate. H_2O_2 solution (30%, 150 mL) was added to digest the organic matter and after standing at room temperature for 24 h, the digested solution was filtered with a vacuum filtration device and additionally filtered through a glass fiber membrane (GF/F, pore size 0.7 μm , diameter 47 mm). Filter membranes were dried at 60 °C.

A total of 13 organism samples (*Bullacta exarata*, *Cerithidea largillierti*, *Macrura veneriformis*, *Sinonovacula constricta*, *Exopalaemon modestus*, *Chinomantes dehaani*, *Scylla serrata*, *Portunus trituberculatus*,

Boleophthalmus pectinirostris, *Uroteuthis edulis*, *Harpadon nehereus*, *Pampus argenteus*, *Larimichthys polyactis*; Table S1) were thawed (−20 °C), washed, and drained. Samples of similar size were selected, body lengths and weight were measured, carapaces shellfish were removed, and the weight was again measured. Tissue digestion was performed using a mixture of 150 mL 10% KOH solution and 50 mL 30% H_2O_2 . The digestion was performed in an oscillatory incubator at 60 °C and 135 rpm/min for at least 24 h until the biological tissue is completely digested where the solution appeared clear. Sediment samples were processed in a similar manner.

Processed samples were observed with a stereomicroscope (SteREO Discovery V8 Zeiss, Carl Zeiss Microimaging GmbH, Germany) and suspected microplastics were identified, photographed (OPLINIC, C33, OPLINIC, America), and counted. The type and color of the microplastics were categorized and the dimensions were measured using ZenLite software (Carl Zeiss Microimaging GmbH, Germany). The chemical components of all suspected microplastics were identified by Fourier-transform infrared spectroscopy (Nicolet iN10, ThermoFisher Scientific, US). Scans were performed 64 times using a 150 $\mu\text{m} \times 150 \mu\text{m}$ aperture at 8 cm^{-1} resolution in liquid nitrogen-cooled transmission mode, and measured spectra were recorded in the 650–4000 cm^{-1} aperture range. Spectra were compared with the organic ensemble spectral library to verify the polymer type. Considering the very small number of microplastic samples at some sites, a matching degree of more than 65% was identified as corresponding to known polymers.

2.4. Statistical methods

The microplastic abundances, sizes, types, and chemical compositions in the sediment and organism samples were characterized based on the experimental results. Sediment microplastic abundance was calculated by weight as items/kg (dw). The microplastic abundance of organisms was calculated by weight as items/g (ww) and by number of individuals as items/individual. The microplastic size was counted every 1000 μm . The microplastic types were divided into six categories: fibers, fragments, lines, films, foams, and pellets.

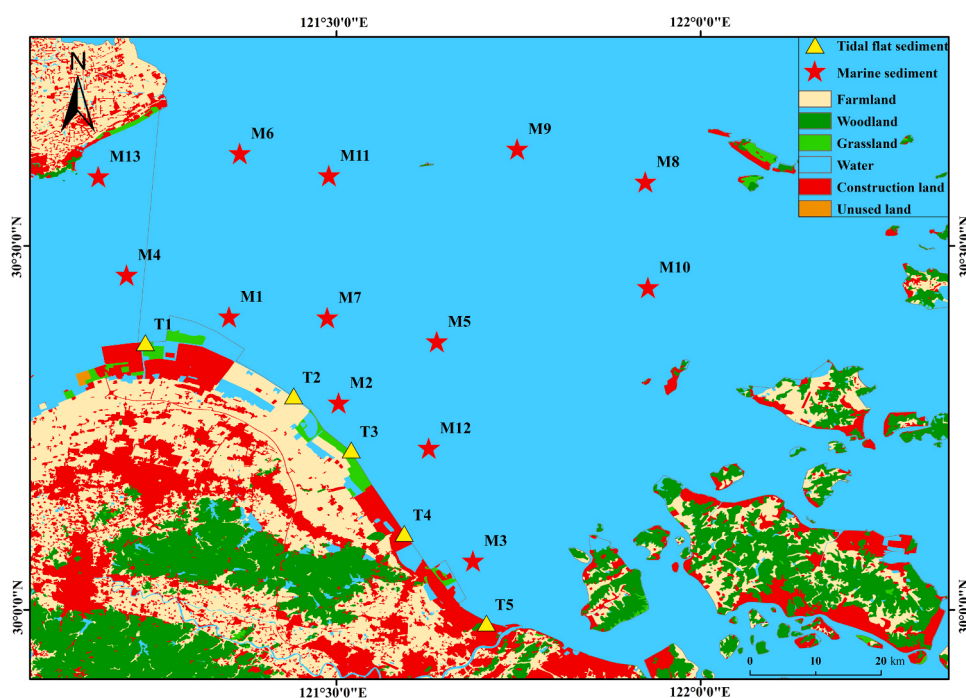


Fig. 1. Map of Hangzhou Bay, showing the sediment sampling locations. Different colors represent different types of land use. ▲: Sampling location; M: Marine; T: Tidal flat.

The method for obtaining the trophic level and specific values can be found in Table S1. Sampling maps were plotted using ArcGIS 10.6, and all resulting maps were completed using Origin 2021 software. One-way analyses of variance, independent sample t-tests, and bivariate correlation analyses were performed on the experimental results using SPSS 19.0 software to test for differences (ANOVA) or correlations (data normally distributed: Pearson correlation analysis; non-normal distribution: Spearman rank correlation coefficient) among the data groups. The term spatial autocorrelation refers to the presence of systematic spatial variation in a mapped variable. Where adjacent observations have similar data values, the map shows positive spatial autocorrelation. Where adjacent observations tend to have very contrasting values, then the map shows negative spatial autocorrelation. A semi-variogram analysis of the spatial distribution of microplastic abundance (using a pure nugget effect model with random variation and distribution of microplastic abundance was performed on microplastic abundance data from two environmental media) was conducted and semi-variogram plotting was completed using GS⁺ Version 9 [27].

2.5. Quality control

Three procedural blank control groups were set up in the experiment, and all the experimental steps of the blank control group were exactly

the same as those of other groups but without experimental samples. All solutions used in the experiment were filtered through glass fiber membranes for later use. No microplastics were found on the glass fiber filter of the blank control group. All glassware and dissection vessels were rinsed three times with filtered Milli-Q water. The utensils used for sample collection and all the tools and containers during the experiment were made of metal or glass. Experimenters wore cotton lab coats and cotton gloves to reduce microplastic pollution in the air while performing field sampling and laboratory work. Samples are stored separately and sealed to prevent any post-collection contamination.

3. Results

3.1. Characteristics of microplastic contamination in sediments of Hangzhou Bay

As shown in Table 1, the overall microplastic abundance in sediments of Hangzhou Bay ranged from 3.32 to 35.12 items/kg, with an average abundance of 16.85 items/kg. Microplastic types were primarily fibers and fragments, black or blue in color, the majority of which were less than 2000 μm ; PP was the main polymer collected. The main polymers in the marine sediments were PL, CE, and RY, which did not match the main polymers of the overall sediments. The microplastic

Table 1
Microplastic contamination in sediments in various environmental media.

Area	Range of abundance (items/kg)	Average abundance (items/kg)	Main types	Main colors	Main size scale (μm)	Main polymers
Marine	3.32–26.15	13.16 $\pm$ 1.82	Fiber (92.16)	Black (52.94)	1–1000 (64.71) 1001–2000 (31.37)	PL (29.41) CE (25.49) RY (23.53)
M1	0.00–9.07	5.88 $\pm$ 2.94	Fiber (100.00)	Black (100)	1–1000 (100.00)	CE (50.00) RY (50.00)
M2	9.17–30.14	19.19 $\pm$ 6.07	Fiber (100.00)	Black (66.67)	1–1000 (100.00)	RY (33.33)
M3	0.00–25.67	8.56 $\pm$ 8.56	Fiber (100.00)	Blue (50.00) White (50.00)	1–1000 (100.00)	PS (50.00) RY (50.00)
M4	0.00–23.68	11.23 $\pm$ 6.86	Fiber (100.00)	Black (75.00)	1001–2000 (50.00)	CE (50.00) PP (50.00)
M5	9.31–34.03	20.12 $\pm$ 7.30	Fiber (100.00)	Blue (40.00) Transparency (40.00)	1–1000 (60.00) 1001–2000 (40.00)	PL (40.00) CE (40.00)
M6	0.00–18.50	9.00 $\pm$ 5.35	Fiber (100.00)	Black (66.67)	1–1000 (33.33) 1001–2000 (33.33) 2001–3000 (33.33)	CE (33.33) CA (33.33) RY (33.33)
M7	0.00–9.95	3.32 $\pm$ 3.32	Fiber (100.00)	White (100.00)	1–1000 (100.00)	PS (100.00)
M8	9.45–30.94	21.28 $\pm$ 6.30	Fiber (83.33)	Black (66.67)	1–1000 (33.33) 1001–2000 (66.67)	PL (50.00) CE (33.33)
M9	0.00–10.57	6.44 $\pm$ 3.26	Fiber (100.00)	Blue (50.00) White (50.00)	1–1000 (50.00) 1001–2000 (50.00)	PL (50.00) RY (50.00)
M10	11.13–48.82	26.15 $\pm$ 11.53	Fiber (75.00)	Black (62.50)	1–1000 (87.50)	PL (37.50) PS (25.00)
M11	10.93–25.72	15.96 $\pm$ 4.88	Fiber (100.00)	Black (75.00)	1–1000 (50.00) 1001–2000 (50.00)	PS (75.00)
M12	0.00–9.53	6.25 $\pm$ 3.12	Fiber (100.00)	Red (50.00) Transparency (50.00)	1–1000 (50.00) 1001–2000 (50.00)	CE (100.00)
M13	9.32–25.66	17.71 $\pm$ 4.72	Fiber (83.33)	Black (50.00) White (33.33)	1–1000 (66.67) 1001–2000 (33.33)	PL (33.33) RY (33.33)
Tidal flat	7.92–35.12	18.63 $\pm$ 2.62	Fiber (52.17) Fragment (29.55)	Black (25.00) Blue (25.00) Green (22.16)	1–1000 (44.89) 1001–2000 (31.25)	PP (38.07)
T1	2.75–32.00	9.43 $\pm$ 4.60	Fiber (68.42)	Black (36.84) Transparency (31.58)	1–1000 31.58) 1001–2000 (52.63)	PP (31.58) PET (21.05)
T2	2.12–16.82	7.92 $\pm$ 4.52	Fiber (100.00)	Blue (30.00) Black (40.00)	1–1000 (40.00) 1001–2000 (30.00)	PP (70.00)
T3	4.39–28.70	14.13 $\pm$ 3.27	Fiber (68.75)	Blue (37.50) Black (28.13)	1–1000 (40.63) 1001–2000 (37.50)	PP (18.75) CE (18.75) RY (18.75)
T4	11.43–34.65	21.19 $\pm$ 3.20	Fiber (40.48) Fragment (38.10)	Green (47.62)	1–1000 (60.47) 1001–2000 (28.57)	PP (54.76)
T5	22.52–52.08	35.12 $\pm$ 5.18	Fiber (41.10) Fragment (39.73)	Blue (23.29) Black (26.03)	1–1000 (41.10) 1001–2000 (24.66) 2001–3000 (20.55)	PP (42.47)
Total	3.32–35.12	16.85 $\pm$ 1.92	Fiber (61.23) Fragment (23.35)	Black (31.28) Blue (21.15)	1–1000 (49.34) 1001–2000 (31.28)	PP (30.40)

Note: The value in brackets is the percentage of each factor in the total number (%).

characteristics of other marine or tidal flat sediments were similar to those of the overall sediments. The legend, abundance, and characteristics of microplastics are shown in Annex Figs. S2–4 and 6.

3.2. Spatial distribution of microplastics in the sediments of Hangzhou Bay

The results of a semi-variance analysis indicated that no spatial distribution pattern was observed in the microplastic abundance from the overall sediments of Hangzhou Bay. However, in the variation model of microplastic abundance in marine sediments, which used a spherical model with spatial dependence, several regions had extremely low or high microplastic abundance (see Fig. 2a). The variation model of microplastic abundance used for tidal flat sediments was a Gaussian model with spatial dependence; in this model, the microplastic abundance was high in the eastern tidal flat section (outlet), but low in the western tidal flat section (inlet) (see Fig. 2b). Annex Fig. S7 shows the semi-variance model of microplastic abundance.

3.3. Characteristics of microplastic contamination in organisms in Hangzhou Bay

Table 2 summarizes the overall microplastic abundance in organisms in Hangzhou Bay, which ranged from 0.04 to 2.71 items/g and from 0.08 to 2.50 items/individual, with average abundances of 0.70 items/g and 0.92 items/individual, respectively. The most common type of

microplastic found was fiber, the main colors were black and blue, the most common sizes were less than 2000 μm , and the main polymers were RY, CE, and PL. The characteristics of microplastics in marine or tidal flat organisms were similar to those in the overall sediments. The abundances and characteristics of microplastics are shown in Annex Figs. S5–6.

The correlation analysis of microplastic abundance performed for each organism and its corresponding trophic level revealed no significant correlation between microplastic abundance and changes in the trophic level of organisms. This result was true when the analysis was applied by both the soft tissue weight and individual counts ($P = 0.239 > 0.05$; $P = 0.492 > 0.05$) (see Fig. 3).

3.4. Correlation of microplastic characteristics between sediments and organisms in Hangzhou Bay

In all microplastics in sediments and organisms, all microplastic indicators, except for color, were correlated in the same environment (Fig. 4). Sediments were significantly correlated with microplastic type within organisms in both the tidal flats and in the overall environment ($P = 0.041 < 0.05$; $P = 0.041 < 0.05$). However, the polymer types in the marine and tidal flat environments were significantly and highly correlated with microplastic polymer types in organisms ($P = 0.010 < 0.05$; $P = 0.001 < 0.01$, respectively). Regardless of the environment, size was highly significantly correlated within both sediments and organisms ($P = 0.001 < 0.01$; $P = 0.005 < 0.01$; $P = 0.001 < 0.01$). A relationship between the microplastic characteristics (type, size, and polymer type) in the organism and those in the environmental media was confirmed.

4. Discussion

4.1. Correlation analysis of microplastic polymer types in sediments and organisms in Hangzhou Bay

The type, color, and size of microplastics change over time due to the physical, chemical, and biological effects of microplastics in the environment. The sources of microplastics in the natural environment cannot be traced by fluorescent labeling. Only the polymer type of microplastics remains the same over time; therefore, polymer type is the best indicator for microplastic source inference or destination analysis.

In total, 17 different microplastic polymer types were detected in sediments and 11 were detected in organisms. Excluding the overlapping polymer types, 21 different microplastic types were detected in sediments and organisms combined (Table S2). A comparison of the proportion of microplastic polymer types in sediments and organisms from the same environment (marine, tidal flats, and overall environment) revealed a significant correlation between microplastics in organisms and those in sediments in marine and tidal flat environments. Likewise, the types of microplastic polymers and number of microplastic samples indicate that the main polymer types and number of samples in sediments and organisms from the same environment are consistent, excluding the singleton (those only occurring once) polymers. Thus, polymer type is the best tracer of the source and transfer of microplastics in the natural environment among the various characteristics of microplastics.

4.2. Comparison of microplastic contamination in organisms in Hangzhou Bay with those from similar bays or tidal flats

The microplastic abundance in marine sediments of Hangzhou Bay in this study was one order of magnitude lower than that in sediments around Chongming Island, Shanghai (12.1 ± 0.9 items/100 g) [28], and the microplastic abundance in tidal flat sediments along the south coast of Hangzhou Bay was slightly lower than that in sediments along the coast of Chongming Island (28.33 items/kg) [29]. Presumably, this

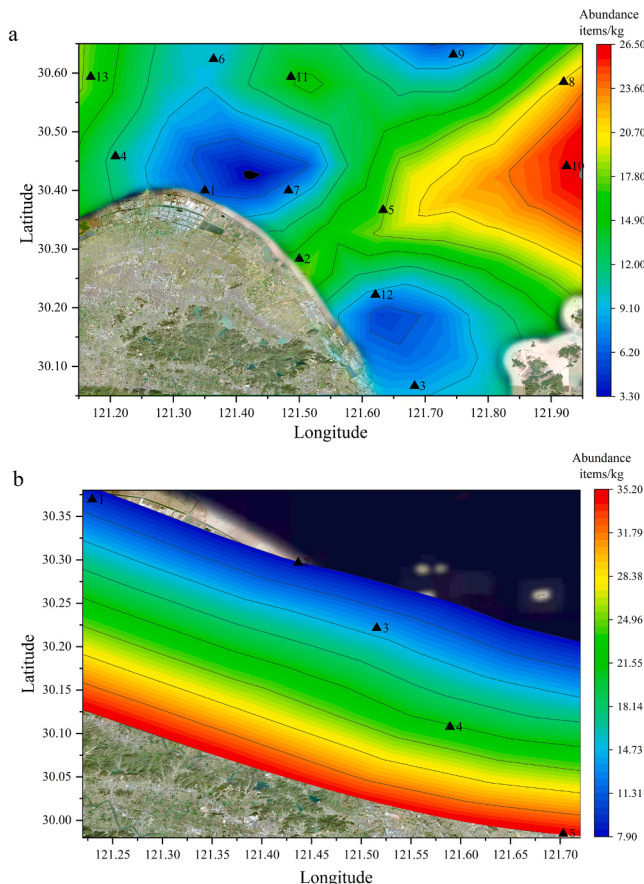


Fig. 2. Fig. 2 Spatial distribution of microplastic abundance in Hangzhou Bay sediments. (a) Marine sediments showing some regions with extremely low or high microplastic abundance as per a spherical model with spatial dependence. (b) Tidal flat sediments depicting a Gaussian model with spatial dependence; high microplastic abundance in the eastern tidal flat section (outlet) and low abundance in the western tidal flat section (inlet). ▲: Sampling location.

Table 2
Microplastic contamination in organisms in various environmental media.

Source	Range of abundance		Average abundance		Main types	Main colors	Main size scale (μm)	Main polymers
	items/g	items/individual	items/g	items/individual				
Marine	0.04–2.71	0.08–2.50	0.62 ± 0.42	0.91 ± 0.37	Fiber (98.04)	Black (54.90) Blue (25.49)	1–1000 (54.90) 1001–2000 (27.45)	PL (33.33) CE (21.57)
<i>Exopalaemon modestus</i>	0.00–20.00	0.00–3.00	2.71 ± 1.88	0.50 ± 0.29	Fiber (83.33)	Black (33.33) Blue (50.00)	1–1000 (66.67)	PL (33.33) PET (33.33)
<i>Portunus triuberculatus</i>	0.00–0.42	0.00–1.00	0.04 ± 0.04	0.08 ± 0.08	Fiber (100.00)	Blue (100.00)	1–1000 (100.00)	PVT (100.00)
<i>Uroteuthis edulis</i>	0.00–0.20	0.00–3.00	0.06 ± 0.03	1.33 ± 0.49	Fiber (100.00)	Blue (50.00)	1–1000 (50.00) 1001–2000 (37.50)	CE (37.50) PET (37.50)
<i>Harpadon nehereus</i>	0.00–2.06	0.00–3.00	0.51 ± 0.32	1.50 ± 0.56	Fiber (100.00)	Black (4.44) Grey (33.33)	1–1000 (44.44) 1001–2000 (33.33)	PL (44.44) PET (33.33)
<i>Pampus argenteus</i>	0.00–0.46	0.00–4.00	0.22 ± 0.09	2.50 ± 0.56	Fiber (100.00)	Black (80.00)	1–1000 (66.67) 1001–2000 (26.67)	CE (26.67) PET (40.00)
<i>Larimichthys polyactis</i>	0.00–0.45	0.00–4.00	0.19 ± 0.07	2.00 ± 0.58	Fiber (100.00)	Black (66.67)	1–1000 (41.67) 1001–2000 (25.00) 2001–3000 (25.00)	PL (25.00) PET (41.67)
Tidal flat	0.08–1.89	0.33–1.67	0.60 ± 0.23	0.83 ± 0.16	Fiber (97.83)	Black (41.30) Blue (43.48)	1–1000 (54.35) 1001–2000 (34.78)	RY (45.65) CE (26.09)
<i>Bullacta exarata</i>	0.00–4.17	0.00–4.00	0.70 ± 0.29	0.83 ± 0.27	Fiber (93.33)	Blue (53.33)	1–1000 (40.00) 1001–2000 (46.67)	RY (33.33) CE (46.67)
<i>Cerithidea largillierti</i>	0.00–1.93	0.00–5.00	0.70 ± 0.32	1.67 ± 0.80	Fiber (100.00)	Black (50.00) Blue (50.00)	1–1000 (60.00) 1001–2000 (30.00)	RY (90.00)
<i>Macraa veneriformis</i>	0.00–1.11	0.00–3.00	0.30 ± 0.18	0.83 ± 0.48	Fiber (100.00)	Black (40.00) Blue (40.00)	1–1000 (80.00)	PET (40.00) CE (40.00)
<i>Sinonovacula constricta</i>	0.00–0.22	0.00–2.00	0.08 ± 0.04	0.67 ± 0.33	Fiber (100.00)	Black (75.00)	1–1000 (25.00) 1001–2000 (25.00)	RY (25.00) PET (25.00)
<i>Chinomantes dehaani</i>	0.00–1.09	0.00–2.00	0.28 ± 0.19	0.50 ± 0.34	Fiber (100.00)	Black (33.33) Grey (33.33) Transparency (33.33)	1–1000 (100.00)	RY (33.33) PET (33.33)
<i>Scylla serrata</i>	0.00–1.01	0.00–1.00	0.24 ± 0.14	0.33 ± 0.17	Fiber (100.00)	Black (67.67)	1–1000 (66.67)	RY (33.33) CE (33.33)
<i>Boleophthalmus pectinirostris</i>	0.00–8.00	0.00–2.00	1.89 ± 1.28	1.00 ± 0.37	Fiber (100.00)	Black (33.33) Blue (50.00)	1–1000 (50.00) 1001–2000 (50.00)	RY (50.00) CE (33.33)
Total	0.04–2.71	0.08–2.50	0.70 ± 0.24	0.92 ± 0.12	Fiber (97.94)	Black (48.45) Blue (34.02)	1–1000 (54.64) 1001–2000 (30.93)	RY (27.84) CE (23.71) PL (20.62)

Note: The value in brackets is the percentage of each factor in the total number (%).

discrepancy is because Chongming Island is part of Shanghai. Although both sites are estuaries along the East China Sea, Chongming Island has more industrial plants and a greater amount of ship traffic, resulting in a greater number microplastics entering the ocean and thus depositing in the sediments. The average microplastic abundance in the marine sediments of Hangzhou Bay was lower than that measured in other bays and estuaries (see Table 3). The average microplastic abundance in the tidal flat sediments of Hangzhou Bay was lower than that in the tidal flat sediments of other bays (see Table 3). The microplastic abundance in the tidal flat sediments of the bay may be closely related to the microplastic abundance in marine sediments (Table 3).

In interpreting the results of this study, it is essential to consider the influence of methodological choices on observed microplastic abundance. Specifically, the use of different floating solutions and sampling depths across studies can introduce variability that might confound comparisons. For instance, different floating solutions, such as NaI, ZnCl₂, and Saturated NaCl, have varying densities, which could potentially affect the buoyancy of microplastics and therefore the detection efficiency [42,43]. Moreover, sediment sampling depths, which ranged from 1 cm to 30 cm in the cited studies, might influence microplastic abundance estimates due to potential stratification of microplastics within sediment layers [44,45]. In shallow sampling depths (e.g., 1 cm), microplastics might be overrepresented due to their potential to be preferentially deposited on the surface layers through low-density or secondary microplastics from the degradation of larger plastic debris [44]. On the other hand, deeper sediment layers (e.g., 30 cm) may contain older microplastics, which have been buried over time [45]. The heterogeneity of methods used across studies underscores the need for

standardization in microplastic research to facilitate more accurate comparisons and to improve our understanding of the true extent of microplastic contamination.

The microplastic abundances in two bivalves in this study were similar to those measured in *Crassostrea gigas* and *Mytilus edulis* (see Table 4), and the microplastic abundance by weight was significantly higher in the *Palaemon modestus* than those in other similar shrimp species. Daniel et al. suggested that the microplastic abundance of *Portunus pelagicus* in the distant sea was the same as that of *Portunus trituberculatus* in this study; both had very low values [46]. The microplastic abundance by weight was extremely low for all mollusks, and the microplastic abundance by individual was greater, probably due to the large specific surface area and body weight of mollusks. Similar to other fishes, the microplastic abundance by individual was greater than the abundance by weight for the four species in this study.

The Xiangshan Bay studied by Wu et al. is close to Hangzhou Bay, but the marine organisms studied are all commercially important species cultured in aquaculture factories in Xiangshan Bay [7], and all the commercially important marine species studied here were native species. Because of the frequent interference of human activities, artificial breeding organisms can be regarded as having great differences compared with those in natural environment. The results of Wu et al. showed that microplastic bioaccumulation was more likely to occur in aquaculture organisms than in organisms in natural environments, which is consistent with the observation that there was no significant correlation between microplastic abundance and trophic level in natural aquatic economic species in this study.

There are many large bay areas around the world, such as the New

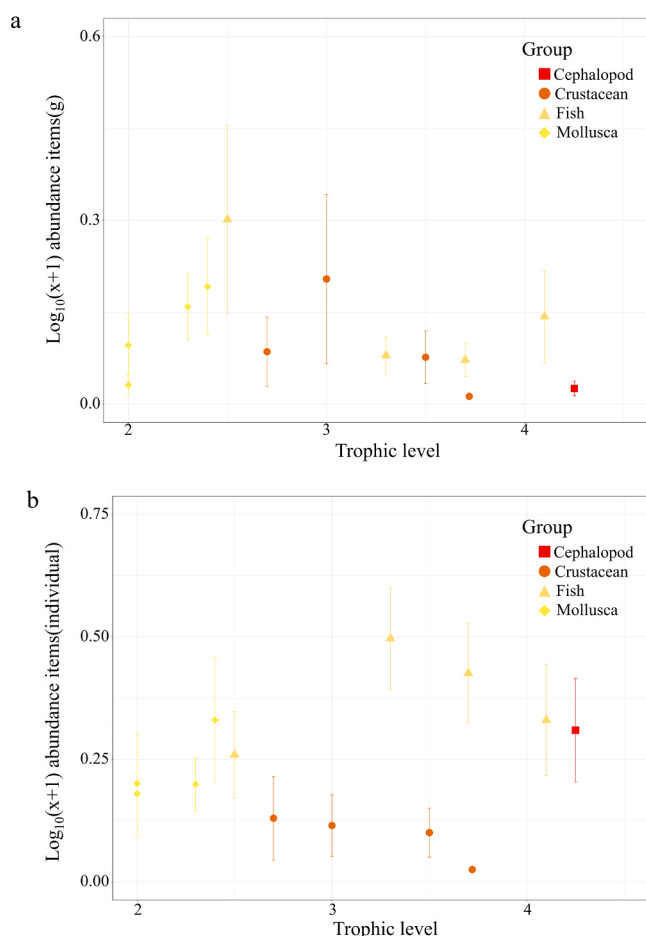


Fig. 3. Relationships between microplastic abundance and trophic level in organisms. No significant correlation was found, regardless of the analysis being based on soft tissue weight (a) or individual counts (b).

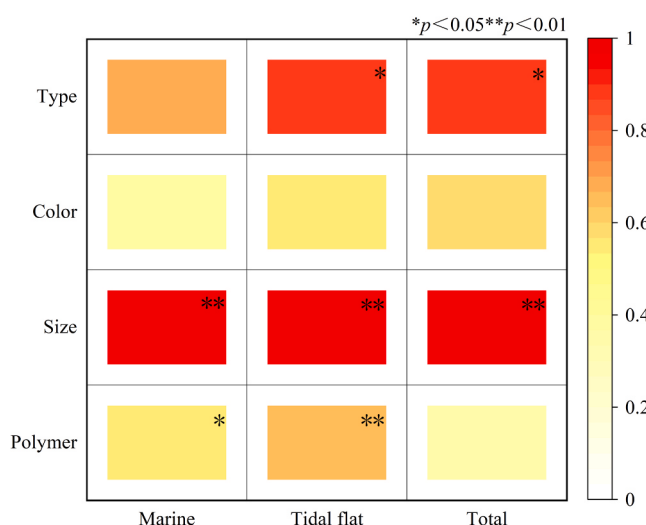


Fig. 4. Correlation analysis of the proportion of microplastic characteristics in sediments and organisms in different environmental media.

York and San Francisco Bay Areas in the United States and the Tokyo Bay Area in Japan [10]. Microplastics in the digestive tracts of 64 Japanese anchovy (*Engraulis japonicus*) sampled in Tokyo Bay were investigated and microplastics were detected in 49 out of 64 fish (77%) [54]. Microplastics transfer between horseshoe crabs (*Limulus polyphemus*)

and migratory shorebirds through consumption of crab eggs was examined in Jamaica Bay, New York, and microplastics were present in all samples [55]. Research on microplastics in San Francisco Bay was among the first studies to conduct a comprehensive and integrated assessment using quality assurance approaches adapted from the field of trace environmental chemistry and microplastic concentrations. The characteristics throughout the bay and surrounding ocean were assessed in both environmental matrices and biota [56]. The present study on microplastic pollution in the Hangzhou Bay Area is intended to catalyze similar efforts to understand and reduce plastic pollution around the globe.

In this study, the amount of microplastic pollution in sediments and in organisms in Hangzhou Bay was similar to that in other bays and organisms of the same kind. The intensity of human activities (e.g., industrial development, ship activity) is the main factor affecting the degree of sediment microplastic pollution. The species, habitat, body size, and trophic level of organisms are the main factors affecting the degree of microplastic pollution in organisms.

4.3. Analysis of microplastic contamination sources in Hangzhou Bay

The polymer PL accounted for the highest proportion in the marine sediments of Hangzhou Bay. PL, commonly known as polyester, is a synthetic fiber commonly used in textiles, and has a high density (1.33 g/cm³), melting point, strength, and chemical stability, as well as strong resistance to light and degradation. Because of this, PL has not easily damaged, which makes it more likely to accumulate in marine sediments compared to other polymers. We assume that marine microplastic contamination mainly comes from incomplete treatment and discharge of laundry wastewater [57].

Polymer PP was found in the highest quantity in the tidal flat sediments of Hangzhou Bay. PP is an organic polymer synthesized by polyaddition reaction. PP has a density between 0.89 and 0.91 g/cm³, and easily floats on the surface of seawater. Furthermore, PP can be utilized within the temperature range of −30–140 °C and can resist corrosion by acid, alkali, and salt at temperature below 80 °C. It is widely used in items closely related to human activities, such as fiber products, fisheries, agriculture, automobiles, bicycles, food, and pharmaceutical packaging. PP materials in the tidal flat zone do not easily enter the ocean with tidal action because their density is lower than that of seawater and because of the high-intensity of human activities in the tidal flats. Therefore, PP microplastics accumulate in large quantities in the tidal flats and are not easily degraded. Thus, PPs remain in large quantities in the tidal flats of Hangzhou Bay.

Among the common microplastic polymer types in organisms in Hangzhou Bay, RY, CE, PL, and PET are widely used in the textile industry and in fisheries (fishing nets and fishing lines). Furthermore, AA can be used to make acrylic plastic, also known as organic glass. The finished plastic products made by the five polymers are colorful; however, it is not yet known whether the color of microplastics affects the feeding efficiency or feeding targets of organisms [19]. The results of this study agreed with the results of a previous study on marine sediments, in which the types of microplastics were dominated by fibers and the proportion of fibers in organisms was extremely high, confirming that microplastic contamination in marine waters is closely related to the discharge of laundry wastewater [45]. Fibrous microplastics that float in seawater also tend to be accidentally ingested by aquatic organisms [46].

In 2020, the highest production of plastic products in China was film (1.50 Mt), accounting for 19.77% of the total. Consequently, in Zhejiang Province, where Hangzhou Bay is located, ranks first in thin film production in China, accounting for 21.73% of total production [58]. Furthermore, Zhejiang Province accounted for 16.84% of China's plastic production in 2020, ranking first in China [58]. However, because the governments of cities along Hangzhou Bay attach great importance to the prevention and control of solid waste pollution, the governments

Table 3

Characteristics of microplastic contamination in sediments in several offshore areas in China.

Study area	Abundance (items/kg)	Main polymer	Main type	Sample depth (cm)	Sampling instrument	Flotation solution	References
Marine sediment							
Xiangshan Bay, China	73.94 ± 30.43	RY, PP, PA	Fiber	5	Van Veen garb sampler	Saturated NaCl	[7]
Yangtze Estuary, China	121.00 ± 9.00	RY, PL, AA	Fiber	5–10	/	Saturated NaCl	[28]
Bohai Sea offshore, China	171.80 ± 55.40	RY, PE, PET	Fiber	/	Stainless steel box sampler	Saturated NaCl	[30]
Northern Yellow Sea offshore, China	123.60 ± 71.60	RY, PE, PET	Fiber	/	Stainless steel box sampler	Saturated NaCl	[30]
Southern Yellow Sea offshore, China	72.00 ± 27.20	RY, PE, PET	Fiber	/	Stainless steel box sampler	Saturated NaCl	[30]
Haizhou Bay, China	330.00 ± 260.00	RY, PET, PP	Fiber	/	Grapple bucket	Saturated NaCl	[31]
Hangzhou Bay, China	13.16 ± 1.82	PL, CE, RY	Fiber	5	Van Veen garb sampler	Saturated NaCl	This study
Chao Phraya River Estuary, Thailand	39.00 ± 14.00	PE, PP	/	/	Van Veen garb sampler	NaI	[32]
Tampa Bay, America	280.00 ± 290.00	/	Fiber	/	Van Dorn sampler	NaI	[33]
Belgian coast, Belgium	390.00	PP, PA	Fiber	/	Van Veen garb sampler	Concentrated saline solution	[34]
Tidal flat sediment							
Beach sand of South China Sea, China	5014.00–8714.00	HDPE, PET, PE	Fiber	1	Watch glass	Saturated NaCl	[35]
Beach sand of Northeastern coast of Beibu Gulf, China	6080.00	HDPE, PET, PE	Fiber	1	Watch glass	Saturated NaCl	[35]
Beach sand of Bohai Sea, China	102.90–163.30	PE, LDPE, HDPE	Fragment	2–20	Metal ring	Saturated NaCl	[36]
Tidal zone of Haizhou Bay, China	490.00 ± 170.00	PE, PET, PS	Fiber	30	Quantitative sampling frame	Saturated NaCl	[9]
Beaches of Xiamen Bay, China	148.27	PE, PS	Fragment	2–3	Stainless steel1 shovel	Saturated NaCl	[37]
Tidal flat of Zhanjiang Bay, China	61.00–989.00 items/m ²	PP	Fragment	2	/	Saturated NaCl	[38]
Hangzhou Bay, China	18.63 ± 2.62	PET, PL, PP+PE	Fiber	5	Stainless steel1 shovel	Saturated NaCl	This study
Kayamkulam estuary, India	433.30	PP	Fiber	/	Van Veen garb sampler	ZnCl ₂	[39]
Vitória bay estuarine system, Brazil	0–126.67	/	Fiber	/	Van Veen garb sampler	Saturated NaCl	[40]
Gulf of Thailand, Thailand	150.40 ± 86.20	RY, PL	Fiber	5	Stainless steel box sampler	Saturated NaCl	[41]

conduct full coverage of solid waste classification, collection, and transportation to improve the utilization rate of resources and develop low-impact treatment methods and standardized management protocols, which results in the relative low abundance of debris and film in sediments of Hangzhou Bay. Regardless, many polymer fibers are too small to be completely filtered and recycled, leading to RY, CE, and PL sediment accumulation, which become the main polymer components of Hangzhou Bay microplastic pollution.

Microplastics were detected in many of the organisms that we examined that are also commonly consumed by people. Although most microplastics are removed through gutting inedible internal organs and cleaning, microplastics are still present in the remaining edible flesh. This consumption includes the microplastics and their adsorbed chemicals and the resulting marine food safety issues and risks to human health require further study.

4.4. Characteristics of microplastic pollution in tidal flat ecosystem of Hangzhou Bay

The overall abundance of microplastics in the sediments of Hangzhou Bay ranged from 3.32 to 35.12 items/kg, but the main types (fiber), colors (black), and sizes (1–1000 µm, 1001–2000 µm) of microplastics were consistent in all environmental media. The predominant type was fiber, was consistent with the results of sediment microplastic pollution in other bays or estuaries around the world (see Table 3). The area of Hangzhou Bay is large and after entering the marine environment, microplastics can be transported for a long distances due to ocean currents and winds. In the process of transportation, microplastics are also

subjected to physical, chemical, and biological effects that will gradually degrade them resulting in microplastics smaller than 2 mm accumulating in the Hangzhou Bay.

The distribution of microplastics in both marine and tidal flat sediments in Hangzhou Bay is spatially dependent. Among benthic sampling sites, M10, M8, and M5 sites are far from the coast and microplastics easily deposit under the action of Lagrange circulation and the net water transport field [59,60], which increases the concentration of microplastics significantly. In contrast, although the sites of M1, M3, M7, and M12 are close to the coast, due to the effect of tides and the channel dredging of Hangzhou Bay in the shipping channel, microplastics are not easily deposited and the concentration of microplastics is reduced. The concentration of microplastics in tidal flat sediments increased slightly from T1 to T2, and then gradually increased to T5 (Fig. S4). On construction land, T4 and T5 experienced a gradual increase of microplastics due to the intensity of human activities, resulting in a very high concentration.

In this study, microplastic pollution was detected in all organisms in Hangzhou Bay. Per weight, *E. modestus* and *B. pectinirostris* had the highest abundance of microplastics (Fig. S5), both of which are omnivorous animals and prefer to inhabit estuarine shoals [61]. Incidentally, the shoal sediments of Hangzhou Bay had a higher abundance of microplastics than those of marine sediments. When counted by individuals, the abundance of microplastics in *P. argenteus* and *L. polyactis* were the highest recorded (Fig. S5). Compared with other organisms in this study, these two species had the highest trophic level and largest body sizes. Furthermore, the adults feed on low-trophic organisms such as jellyfish, benthic animals, and small fish [61], leading to

Table 4
Characteristics of microplastic contamination in several marine organisms.

Species	Site	Abundance		Main Type	References
		Items/g	Items/ individual		
Bivalve, Snail					
<i>Ostrea denselamellosa</i>	Xiangshan Bay	0.31 ± 0.10	1.67 ± 0.44	Fiber	[7]
<i>Sinonovacula constricta</i>	Xiangshan Bay	0.21 ± 0.05	1.80 ± 0.34	Fiber	[7]
<i>Mytilus edulis</i>	A mussel farm in Germany	0.36 ± 0.07	/	/	[47]
<i>Crassostrea gigas</i>	Purchase from supermarket	0.47 ± 0.16	/	/	[47]
<i>Modiolus modiolus</i>	Coast of Scotland	0.086 ± 0.031	3.50 ± 1.29	Fiber	[48]
<i>Bullacta exarata</i>	Hangzhou Bay	0.70 ± 0.29	0.83 ± 0.27	Fiber	This study
<i>Cerithidea largillierti</i>	Hangzhou Bay	0.70 ± 0.32	1.67 ± 0.80	Fiber	This study
<i>Macraa veneriformis</i>	Hangzhou Bay	0.30 ± 0.18	0.83 ± 0.48	Fiber	This study
<i>Sinonovacula constricta</i>	Hangzhou Bay	0.08 ± 0.04	0.67 ± 0.33	Fiber	This study
Shrimp					
<i>Parapenaeopsis hardwickii</i>	Xiangshan Bay	0.25 ± 0.08	0.95 ± 0.28	Fiber	[7]
<i>Lysmata vittata</i>	Xiangshan Bay	/	0.30 ± 0.50	Fiber	[49]
<i>Exopalaemon modestus</i>	Hangzhou Bay	2.71 ± 1.88	0.50 ± 0.29	Fiber	This study
Crab					
<i>Portunus pelagicus</i>	fishing harbors of India	0.003 ± 0.01	0.14 ± 0.44	Fragment	[46]
Coastal sea crab	Zhoushan	2.37 ± 0.41	/	Fiber	[50]
<i>Chinomantes dehaani</i>	Hangzhou Bay	0.28 ± 0.19	0.50 ± 0.34	Fiber	This study
<i>Scylla serrata</i>	Hangzhou Bay	0.24 ± 0.14	0.33 ± 0.17	Fiber	This study
<i>Portunus trituberculatus</i>	Hangzhou Bay	0.04 ± 0.04	0.08 ± 0.08	Fiber	This study
Mollusc					
<i>Sepia officinalis</i>	Marine station, Portugal	/	39.00	Fiber	[51]
<i>Uroteuthis (Photololigo) duvaucelii</i>	fishing harbors of Kerala, India	0.008 ± 0.02	0.18 ± 0.48	Fragment	[46]
<i>Dosidicus gigas</i>	The coast of Peru	0.625	5.950	Fiber	[52]
<i>Uroteuthis edulis</i>	Hangzhou Bay	0.06 ± 0.03	1.33 ± 0.49	Fiber	This study
Fish					
<i>Konosirus punctatus</i>	Xiangshan Bay	0.044 ± 0.025	2.1 ± 0.38	Fiber	[7]
<i>Larimichthys crocea</i>	Xiangshan Bay	0.008 ± 0.006	1.80 ± 0.42	Fiber	[7]
<i>Myctophum aulolaternatum</i>	North Pacific	/	6.00 ± 8.99	Fragment	[53]
	Central Gyre				
<i>Cololabis saira</i>	North Pacific	/	3.20 ± 3.05	Fragment	[53]
	Central Gyre				
Planktivorous fish	Tokyo Bay	/	2.30 ± 2.50	Fragment	[54]
<i>Boleophthalmus pectinirostris</i>	Hangzhou Bay	1.89 ± 1.28	1.00 ± 0.37	Fiber	This study
<i>Harpadon nehereus</i>	Hangzhou Bay	0.51 ± 0.32	1.50 ± 0.56	Fiber	This study
<i>Pampus argenteus</i>	Hangzhou Bay	0.22 ± 0.09	2.50 ± 0.56	Fiber	This study
<i>Larimichthys polyactis</i>	Hangzhou Bay	0.19 ± 0.07	2.00 ± 0.58	Fiber	This study

microplastic bioaccumulation. Hypothetically, microplastics pass through the food web and accumulate in higher level trophic organisms. However, we observed no significant correlation between the abundance of microplastics in organisms and trophic level (Fig. 3). This unexpected finding may be related to the large species differences among organisms in this study. In the future, more similar species groupings should be selected for analysis.

4.5. Risk prediction of microplastic pollution in marine organisms in Hangzhou Bay

In 2020, China's total marine fishery output reached 10.94 M t, among which Zhejiang province, where Hangzhou Bay is located, had a marine fishery output of 2.57 M t, ranking first in China [62]. China is bordered by the Bohai Sea, the Yellow Sea, the East China Sea, and the South China Sea. Hangzhou Bay is the mouth of the Yangtze River entrance into the East China Sea. The total output of marine fisheries in 2020 in the East China Sea was 3.81 M t, while Zhejiang's marine fishery output in the East China Sea was 2.39 M t, the largest in China. In 2020, aquatic products in Zhejiang province were exported as a total of 172.76 M USD and 0.44 M t, mostly to the United States, the European Union, the Russian Federation, South Korea, Japan, Thailand, and other countries and regions. Therefore, distribution of microplastic contaminants in seafood from Zhejiang may impact global trading partners.

In 2020, 1.72 M t of fish, 0.50 M t of shrimp, 0.18 M t of crabs, 20.50 k t of shellfish and 0.13 M t of cephalopods were caught in the waters off Zhejiang province [62]. Among the representative marine economic species selected in this study, *Boleophthalmus pectinirostris*, *Harpadon nehereus*, *Pampus argenteus*, and *Larimichthys polyactis* are fish,

Exopalaemon modestus are shrimp, *Chinomantes dehaani*, *Scylla serrata*, and *Portunus trituberculatus* are crabs, *Bullacta exarata*, *Cerithidea largillierti*, *Macraa veneriformis*, *Sinonovacula constricta* and *Uroteuthis edulis* are mollusks. Based on the results of this study, it can be inferred that in 2020, the total amount of these five types of marine economic species caught in Zhejiang province could accumulate about 2.23 G t of microplastic particles. Thus, the potential risks to human health caused by microplastic pollution of marine organisms cannot be ignored.

3. Outlook and conclusion

This study confirmed the existence of high microplastic pollution in marine sediments, tidal flat sediments, and 13 commercially important species in the Hangzhou Bay. However, the marine ecosystem of Hangzhou Bay is not only composed of sediments and organisms, and the 13 aquatic economic species cannot fully represent the marine organisms and complex food web of Hangzhou Bay. As such, the existing research results have not been able to provide a detailed answer to the various microplastics pollution problems in the marine ecosystem of Hangzhou Bay and its marine organisms. Future studies should be further conducted considering the following aspects: (1) How microplastics get transferred in the marine environment; (2) Quantitative tracing analysis of microplastics to assess the range of microplastic types and accumulation in marine ecosystems.

High microplastic contamination was found in the tidal flats and organisms in Hangzhou Bay (average abundance of sediments: 16.85 ± 1.92 items/kg; average abundance of organisms: 0.70 ± 0.24 items/g, 0.92 ± 0.12 items/individual), and several microplastic characteristics in sediments and organisms were correlated in the same environment

(type, size, and polymer type), confirming the link between microplastic contamination in organisms and the environment. Furthermore, we found (1) spatial autocorrelation in the distribution of microplastic abundances in marine and tidal flat sediments in Hangzhou Bay, but no spatial autocorrelation in the offshore tidal flat environment as a whole. (2) The microplastic abundance in each organism was not significantly correlated with its corresponding trophic level, either by weight or by individual ($P = 0.239 > 0.05$; $P = 0.492 > 0.05$, respectively). (3) This study has demonstrated that microplastics present in marine and tidal flat ecosystems are transferred from the environment to the network of marine organisms and finally to humans through the food web, which poses a threat to human life, health, and safety. In summary, the situation of microplastic pollution in Hangzhou Bay is relatively widespread. It is necessary to formulate management plans to effectively control its spread, and to reduce the harm caused by microplastic pollution to marine organisms in Hangzhou Bay and reduce the threat to human health.

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CRediT authorship contribution statement

Xinyi Cai: Conceptualization, Methodology, Investigation, Writing – original draft, Software, Statistical analysis, Writing – review & editing. **Huili Chen:** Conceptualization, Methodology, Investigation, Writing – original draft, Supervision, Writing – review & editing. **Jie Cheng:** Investigation, Writing – original draft, Software, Statistical analysis. **Bei Huang:** Investigation, Writing – original draft. **Binsong Jin:** Conceptualization, Methodology, Supervision, Writing – review & editing. **Jianbo Lu:** Conceptualization, Methodology, Investigation, Writing – original draft, Supervision, Writing – review & editing. All authors gave the final approval for publication.

Environmental Implication

The pollution caused by microplastics and the new health threats they pose to all mankind are urgent ecological and public health issues that need to be addressed. This study reveals significant microplastic contamination in sediments and organisms in Hangzhou Bay China. The microplastics are spatially distributed across the bay with no correlation with organisms' trophic levels. The findings highlight that microplastics in organisms likely originate from the sediments they inhabit. These results underscore the environmental implications of microplastic pollution, affecting marine life, potentially carrying toxins, and showing the need for targeted cleanup efforts and strategies to reduce microplastic pollution sources.

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.

Data Availability

Data will be made available on request.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.jhazmat.2023.131838](https://doi.org/10.1016/j.jhazmat.2023.131838).

References

- [1] Law, K.L., Thompson, R.C., 2014. Microplastic in the seas. *Science* 345, 144–145. <https://doi.org/10.1126/science.1254065>.
- [2] Andrady, A.L., 2011. Microplastics in the marine environment. *Mar Pollut Bull* 62. <https://doi.org/10.1016/j.marpolbul.2011.05.030>.
- [3] Leslie, H.A., Brandsma, S.H., van Velzen, M.J.M., Vethaak, A.D., 2017. Microplastics en route: Field measurements in the Dutch river delta and Amsterdam canals, wastewater treatment plants, North Sea sediments and biota. *Environ Int* 101, 133–142. <https://doi.org/10.1016/j.envint.2017.01.018>.
- [4] Jambeck, J.R., Geyer, R., Wilcox, C., Siegler, T.R., Perryman, M., Andrady, A., Narayan, R.K., Law, L., 2015. Plastic waste inputs from land into the ocean. *Science* 347, 768–771. <https://doi.org/10.1126/science.1260352>.
- [5] Meijer, L.J.J., van Emmerik, T., van der Ent, R., Schmidt, C., Lebreton, L., 2021. More than 1000 rivers account for 80% of global riverine plastic emissions into the ocean. *Sci Adv* 7, eaaz5803. <https://doi.org/10.1126/sciadv.aaz5803>.
- [6] Garcia-Chamero, A., Hernandez, C.M.A., Lago, D.C., 2020. First evidence of microplastics in Cienfuegos bay, Cuba. *Ecosistemas* 29, 2085. <https://doi.org/10.7818/ECOS.2085>.
- [7] Wu, F., Wang, Y., Leung, J., Huang, W., Zeng, J., Tang, Y., Chen, J., Shi, A., Yu, X., Xu, X., Zhang, H., Cao, L., 2020. Accumulation of microplastics in typical commercial aquatic species: A case study at a productive aquaculture site in China. *Sci Total Environ* 708, 135432. <https://doi.org/10.1016/j.scitotenv.2019.135432>.
- [8] Taha, Z.D., Amin, R.M., Anuar, S.T., Nasser, A.A.A., Sohaimi, E.S., 2021. Microplastics in seawater and zooplankton: a case study from Terengganu estuary and offshore waters, Malaysia. *Sci Total Environ* 786, 147466. <https://doi.org/10.1016/j.scitotenv.2021.147466>.
- [9] Wang, J., Song, K., Sun, Y., Fang, T., Li, J., Zhang, T., Feng, Z., 2021. Indicator function of ragworm (*Nereididae*) on sediment microplastic in Haizhou Bay intertidal zone. *Environ Sci* 42, 4341–4349. <https://doi.org/10.13227/j.hjlx.202012069>.
- [10] Cai, W., Han, B., Lu, F., Xian, C., Ouyang, Z., 2020. Comprehensive evaluation of the eco-environment in the four global bay areas. *Acta Ecol Sin* 40, 8392–8402. <https://doi.org/10.5846/stxb202004301062>.
- [11] Fok, L., Cheung, P.K., 2015. Hong Kong at the pearl river estuary: a hotspot of microplastic pollution. *Mar Pollut Bull* 99, 112–118. <https://doi.org/10.1016/j.marpolbul.2015.07.050>.
- [12] Zhao, S., Zhu, L., Li, D., 2015. Microplastic in three urban estuaries, China. *Environ Pollut* 206, 597–604. <https://doi.org/10.1016/j.envpol.2015.08.027>.
- [13] Retama, I., Jonathan, M.P., Shruti, V.C., Velumani, S., Sarkar, S.K., Roy, P.D., Rodríguez-Espinosa, P.P., 2016. Microplastics in tourist beaches of Huatulco Bay, Pacific coast of southern Mexico. *Mar Pollut Bull* 113, 530–535. <https://doi.org/10.1016/j.marpolbul.2016.08.053>.
- [14] Nakano, H., Arakawa, H., Tokai, T., 2021. Microplastics on the sea surface of the semi-closed Tokyo Bay. *Mar Pollut Bull* 162, 111887. <https://doi.org/10.1016/j.marpolbul.2020.111887>.
- [15] Cai, X., Chen, H., Huang, B., Lu, J., 2022. Analysis on advances and characteristics of microplastic pollution in China's lake ecosystems. *Ecotox Environ Safe* 232, 113254. <https://doi.org/10.1016/j.ecoenv.2022.113254>.
- [16] Ruangpanupan, N., Ussawarujikulchai, A., Prapagdee, B., Suchana, C., 2022. Microplastics in the surface seawater of Bandon Bay, Gulf of Thailand. *Mar Pollut Bull* 179, 113664. <https://doi.org/10.1016/j.marpolbul.2022.113664>.
- [17] Carbery, M., O'Connor, W., Palanisami, T., 2018. Trophic transfer of microplastics and mixed contaminants in the marine food web and implications for human health. *Environ Int* 115, 400–409. <https://doi.org/10.1016/j.envint.2018.03.007>.
- [18] Li, C., Gan, Y., Zhang, C., He, H., Fang, J., Wang, L., Wang, Y., Liu, J., 2021. "Microplastic communities" in different environments: differences, links, and role of diversity index in source analysis. *Water Res* 188, 116574. <https://doi.org/10.1016/j.watres.2020.116574>.
- [19] Hossain, M., Olden, J., 2022. Global meta-analysis reveals diverse effects of microplastics on freshwater and marine fishes. *Fish Fish* 23, 1439–1454. <https://doi.org/10.1111/faf.12701>.
- [20] Adeogun, A.O., Ibor, O.R., Khan, E.A., Chukwuka, A.V., Omogbemi, E.D., Arukwe, A., Detection and occurrence of microplastics in the stomach of commercial fish species from a municipal water supply lake in southwestern Nigeria. *Environ Sci Pollut R* 27, 31035–31045. <https://doi.org/10.1007/s11356-020-09031-5>.
- [21] Sfriso, A.A., Tomio, Y., Rosso, B., Gambaro, A., Sfriso, A., Corami, F., Rastelli, E., Corinaldesi, C., Mistri, M., Munari, C., 2020. Microplastic accumulation in benthic invertebrates in Terra Nova Bay (Ross Sea, Antarctica). *Environ Int* 137, 105587. <https://doi.org/10.1016/j.envint.2020.105587>.
- [22] Frimpong-Manso, J.P., Vivar, I., 2019. Trophic transfer of microplastics in aquatic ecosystems (Presentation). <https://doi.org/10.13140/RG.2.2.11637.32481>.
- [23] Bour, A., Avio, C.G., Gorb, S., Regoli, F., Hylland, K., 2018. Presence of microplastics in benthic and epibenthic organisms: Influence of habitat, feeding mode and trophic level. *Environ Pollut* 243 (Pt B), 1217–1225. <https://doi.org/10.1016/j.envpol.2018.09.115>.
- [24] Li, L., Jiang, M., Wang, Y., 2018. *Aquatic Biological Resources and Environment in Hangzhou Bay*. China Agriculture Press, Beijing.

- [25] Pang, H., Lyu, S., Chen, X., Jin, A., Loh, P., Li, F., Jiang, Y., Yang, X., Yan, K., Lou, Z., 2017. Heavy metal distribution and accumulation in the *Spartina alterniflora* from the Andong tidal flat, Hangzhou Bay, China. *Environ Earth Sci* 76, 627–641. <https://doi.org/10.1007/s12665-017-6948-3>.
- [26] Sun, X., Li, Q., Zhu, M., Liang, J., Zheng, S., Zhao, Y., 2016. Ingestion of microplastics by natural zooplankton groups in the northern South China Sea. *Mar Pollut Bull* 115, 217–224. <https://doi.org/10.1016/j.marpolbul.2016.12.004>.
- [27] Zheng, X., Lv, L., 2018. *Geostatistics (Modern Spatial Statistics)*. China Science Press, Beijing.
- [28] Peng, G., Zhu, B., Yang, D., Su, L., Shi, H., Li, D., 2017. Microplastics in sediments of the Changjiang Estuary, China. *Environ Pollut* 225, 283–290. <https://doi.org/10.1016/j.envpol.2016.12.064>.
- [29] Li, Y., Lu, Z., Zheng, H., Wang, J., Chen, C., 2020. Microplastics in surface water and sediments of Chongming Island in the Yangtze Estuary, China. *Environ Sci Eur* 32, 15. <https://doi.org/10.1186/s12302-020-0297-7>.
- [30] Zhao, J., Ran, W., Teng, J., Liu, Y., Liu, H., Yin, X., Cao, R., Wang, Q., 2018. Microplastic pollution in sediments from the Bohai Sea and the Yellow Sea, China. *Sci Total Environ* 640–641, 637–645. <https://doi.org/10.1016/j.scitotenv.2018.05.346>.
- [31] Li, Z., Gao, C., Yang, J., Wu, L., Zhang, S., Liu, Y., Jin, D., 2020. Distribution characteristics of microplastics in surface water and sediments of Haizhou Bay, Lianyungang. *Environ Sci* 41, 3212–3221. <https://doi.org/10.13227/j.hjkk.201910005>.
- [32] Ta, A.T., Babel, S., 2020. Microplastics pollution with heavy metals in the aquaculture zone of the Chao Phraya River Estuary, Thailand. *Mar Pollut Bull* 161 (Pt A), 111747. <https://doi.org/10.1016/j.marpolbul.2020.111747>.
- [33] McEachern, K., Alegria, H., Kalagher, A.L., Hansen, C., Morrison, S., Hastings, D., 2019. Microplastics in Tampa Bay, Florida: abundance and variability in estuarine waters and sediments. *Mar Pollut Bull* 148, 97–106. <https://doi.org/10.1016/j.marpolbul.2019.07.068>.
- [34] Claessens, M., Meester, S., Landuyt, De, Van, L., Clerck, K.De, Janssen, C.R., 2011. Occurrence and distribution of microplastics in marine sediments along the Belgian coast. *Mar Pollut Bull* 62 (10), 2199–2204. <https://doi.org/10.1016/j.marpolbul.2011.06.030>.
- [35] Qiu, Q., Peng, J., Yu, X., Chen, F., Wang, J., Dong, F., 2015. Occurrence of microplastics in the coastal marine environment: first observation on sediment of China. *Mar Pollut Bull* 98, 274–280. <https://doi.org/10.1016/j.marpolbul.2015.07.028>.
- [36] Yu, X., Peng, J., Wang, J., Wang, K., Bao, S., 2016. Occurrence of microplastics in the beach sand of the Chinese inner sea: the Bohai Sea. *Environ Pollut* 412, 722–730. <https://doi.org/10.1016/j.envpol.2016.04.080>.
- [37] Liu, Q., Liang, H., Xi, G., Hu, X., Ge, J., 2019. Microplastic Pollution of the Beaches in Xiamen Bay, China. *Environ Sci* 40, 1217–1221. <https://doi.org/10.13227/j.hjkk.201807217>.
- [38] Yao, S., Lin, Z., Feng, M., Qiu, Z., Hu, Y., Chen, F., 2021. Microplastic pollution of the beaches in Zhanjiang Bay, China. *Guangdong Chem Ind* 48, 60–64. <https://doi.org/10.3969/j.issn.1007-1865.2021.15.023>.
- [39] Radhakrishnan, K., Sivapriya, V., Rajkumar, A., Akramkhan, N., Prakasheswar, P., Krishnakumar, S., Hussain, S.M., 2021. Characterization and distribution of microplastics in estuarine surface sediments, Kayamkulam estuary, southwest coast of India. *Mar Pollut Bull* 168, 112389. <https://doi.org/10.1016/j.marpolbul.2021.112389>.
- [40] Neto, J.A.B., Gaylarde, C., Beech, I., Bastos, A.C., Quaresma, V.D.S., de Carvalho, D.G., 2019. Microplastics and attached microorganisms in sediments of the Vitória bay estuarine system in SE Brazil. *Ocean Coast Manag* 169, 247–253. <https://doi.org/10.1016/j.ocecoaman.2018.12.030>.
- [41] Wang, Y., Zou, X.Q., Peng, C., Qiao, S.Q., Wang, T., Yu, W.W., Khokiatwong, O. A., Kornkanitnan, N., 2020. Occurrence and distribution of microplastics in surface sediments from the Gulf of Thailand. *Mar Pollut Bull* 152, 110916. <https://doi.org/10.1016/j.marpolbul.2020.110916>.
- [42] Hidalgo-Ruz, V., Gutow, L., Thompson, R.C., Thiel, M., 2012. Microplastics in the marine environment: a review of the methods used for identification and quantification. *Environ Sci Technol* 46 (6), 3060–3075. <https://doi.org/10.1021/es2031505>.
- [43] Löder, M.G., Gerdts, G., 2015. *Methodology used for the detection and identification of microplastics—a critical appraisal*. *Marine Anthropogenic Litter*. Springer, Cham, pp. 201–227. https://doi.org/10.1007/978-3-319-16510-3_8.
- [44] Fischer, E.K., Paglialonga, L., Czech, E., Tamminga, M., 2016. Microplastic pollution in lakes and lake shoreline sediments – a case study on Lake Bolsena and Lake Chiusi (central Italy). *Environ Pollut* 213, 648–657. <https://doi.org/10.1016/j.envpol.2016.03.012>.
- [45] Vianello, A., Boldrin, A., Guerriero, P., Moschino, V., Rella, R., Sturaro, A., Da Ros, L., 2013. Microplastic particles in sediments of Lagoon of Venice, Italy: First observations on occurrence, spatial patterns and identification. *Estuar Coast Shelf Sci* 130, 54–61. <https://doi.org/10.1016/j.ecss.2013.03.022>.
- [46] Daniel, D., Ashraf, B.P.M., Thomas, S.N., Thomson, K.T., 2021. Microplastics in the edible tissues of shellfishes sold for human consumption. *Chemosphere* 264, 128554. <https://doi.org/10.1016/j.chemosphere.2020.128554>.
- [47] Van Cauwenberghe, L., Janssen, C.R., 2014. Microplastics in bivalves cultured for human consumption. *Environ Pollut* 193, 65–70. <https://doi.org/10.1016/j.envpol.2014.06.010>.
- [48] Catarino, A.I., Macchia, V., Sanderson, W.G., Thompson, R.C., Henry, T.B., 2018. Low levels of microplastics (MP) in wild mussels indicate that MP ingestion by humans is minimal compared to exposure via household fibres fallout during a meal. *Environ Pollut* 237, 675–684. <https://doi.org/10.1016/j.envpol.2018.02.069>.
- [49] Yu, X., Xu, L., Liu, Q., Huang, W., Cao, L., Dou, S., 2021. Abundance, shape, and chemical composition of microplastics within ten common marine species in Xiangshan Bay. *Prog Fish Sci* 42, 9–19. <https://doi.org/10.19663/j.issn2095-9869.20200221002>.
- [50] Wang, H., Zhao, H., Gui, F., Zhao, S., 2019. Characteristics of microplastic pollution in sea crabs in coastal waters of Zhoushan. *Rural Econ Sci-Technol* 30, 66–68.
- [51] Oliveira, A.R., Sardinha-Silva, A., Andrews, L.R.P., Green, D., Cooke, G.M., Hall, S., Blackburn, K., Sykes, A.V., 2020. Microplastics presence in cultured and wild-caught cuttlefish, *Sepia officinalis*. *Mar Pollut Bull* 160, 111553. <https://doi.org/10.1016/j.marpolbul.2020.111553>.
- [52] Wang, Y., Fan, J., Gong, Y., Niu, C., Chen, X., 2022. Microplastics in the stomach and intestine of pelagic squid: a case study of *Dosidicus gigas* in open sea of Peru. *Chin J Appl Ecol* 33, 255–260. <https://doi.org/10.13287/j.1001-9332.202201.038>.
- [53] Boerger, C.M., Lattin, G.L., Moore, S.L., Moore, C.J., 2010. Plastic ingestion by planktivorous fishes in the North Pacific Central Gyre. *Mar Pollut Bull* 60, 2275–2278. <https://doi.org/10.1016/j.marpolbul.2010.08.007>.
- [54] Tanaka, K., Takada, H., 2016. Microplastic fragments and microbeads in digestive tracts of planktivorous fish from urban coastal waters. *Sci Rep* 6, 34351. <https://doi.org/10.1038/srep34351>.
- [55] Ward, R.M., Casper, E.M., Clark, J.A., Botton, M.L., 2022. Microplastic transfer from the American horseshoe crab to shorebirds through consumption of horseshoe crab eggs in Jamaica Bay, NY. *Mar Pollut Bull* 184, 114148. <https://doi.org/10.1016/j.marpolbul.2022.114148>.
- [56] Miller, E., Sedlak, M., Lin, D., Box, C., Holleman, C., Rochman, C.M., Sutton, R., 2021. Recommended best practices for collecting, analyzing, and reporting microplastics in environmental media: lessons learned from comprehensive monitoring of San Francisco Bay. *J Hazard Mater* 409 (9), 124770. <https://doi.org/10.1016/j.jhazmat.2020.124770>.
- [57] Browne, M.A., Crump, P., Niven, S.J., Teuten, E., Tonkin, A., Galloway, T., Thompson, R., 2011. Accumulation of microplastic on shorelines worldwide: Sources and sinks. *Environ Sci Technol* 45, 9175–9179. <https://doi.org/10.1021/es201811s>.
- [58] China Plastics Processing Industry Association, 2022. *China Plastics Industry Yearbook 2021*. Chinese Light Industry Press, Beijing.
- [59] Zhou, T., 2008. Numerical simulation of lagrange residual current in the Changjiang Estuary, Hangzhou Bay and their adjacent sea. *Ocean University of China*.
- [60] Pan, C.H., Hu, G.J., Tang, Z.W., You, A.J., Lu, H.Y., Cao, Y., Yu, P.B., Xie, D.F., Ding, T., Zhu, J.Z., He, R.Y., 2010. Regulation and development of Qiantang River estuary—study on regular pattern of water and sediment movement and riverbed evolution. *Apprais Doc Sci Technol Proj Zhejiang Prov Dep Sci Technol*. (<https://kns.cnki.net/KCMS/detail/detail.aspx?dbname=SNAD&filename=SNAD000001413858>).
- [61] Zhao, S.L., Xu, H.X., Zhong, J.S., Xu, J., 2017. *Marine Fish Record of Zhejiang Province*. Zhejiang Science and Technology Press, Hangzhou.
- [62] Ministry of Agriculture and Rural Affairs of China, 2021. *China Fishery Statistical Yearbook 2020*. China Agriculture Press, Beijing.