



Original Research

Unveiling microplastic distribution and interactions in the benthic layer of the Yangtze River Estuary and East China Sea



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ABSTRACT

Microplastics (MPs), recognized as an emerging global environmental concern, have been extensively detected worldwide, with specific attention directed towards the Yangtze River Estuary (YRE) and East China Sea (ECS) regions. Despite their critical research significance, there remains a knowledge gap concerning the distribution of MPs in the benthic layer within this area, particularly regarding interactions governing their occurrence. Here we illuminate the distribution of MPs within the benthic layer and unravel the intricate interplay between bottom water and sediment in the YRE and ECS. We find that MPs are notably more abundant in bottom water, ranging from 8 to 175 times higher than in surface water. These MPs predominantly consist of polyester fibers, exhibit a size range between 0.5 and 5.0 mm, and display distinct coloration. Co-occurrence network analysis and Principal Coordinate Analysis confirm a robust correlation between MPs in bottom water and sediment, signifying the pivotal role of bottom water in mediating the distribution and transportation of MPs within the benthic layer. Furthermore, a positive correlation between MPs in sediment and bottom water turbidity underscores the impact of surface sediment resuspension and upwelling on MPs distribution. This study clarifies the intricate interactions within the benthic layer and highlights the crucial role of bottom water as a mediator in the vertical distribution of MPs, advancing our understanding of the “source-to-sink” transport processes governing MPs within water-sediment systems.

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1. Introduction

Increasing global attention has been paid to the occurrence and distribution of microplastics (MPs) due to their vast presence in the atmosphere, surface water, sediments, soils, and even many organisms [1–5]. The vertical distribution of MPs in water has been extensively studied [6–10], with the significant findings that the MPs abundances in surface and intermediate waters were significantly lower than that of bottom water [7,8]. For instance, Lima et al. [7] found that the abundance of MPs in the bottom water of

the Goiana Estuary is 1.53 items L⁻¹, several times higher than those in the surface water (0.35 and 0.43 items L⁻¹). The MPs can further sink and eventually deposit in river and marine sediment [11,12]. The surface sediment was also recognized as the source of pollutants, and the MPs in the sediments may be potentially transferred to the bottom water together with sediment particle resuspension [8,13]. Additionally, the toxic effects of MPs on marine organisms in the bottom water and sediment, especially for the bottom dwellers, have been widely reported [3,4]. The bottom water and sediment are closely connected as a whole system and could mutually affect the MPs distribution. However, there remains a research gap regarding the interactions of MPs at the water-sediment interface [2,6,8,10,13].

Currently, many studies have been reported on the distributions of MPs in river and marine environments, as well as delved into the vertical profiles of MPs in water and sediment [8,10,14]. In general,

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the distribution of MPs exhibits marked variations across distinct sampling stations, primarily attributable to factors such as point source discharge, biological activities, and MPs transportation [9]. More abundant MPs were found in the most urbanized sites, sewage discharges, and near the maximum turbidity front [15]. Besides, the size, shape, and density of MPs may affect their vertical distribution in water and sediment via floatation, sinking to the bottom, or resuspension in the water column [16,17]. Moreover, many environmental parameters, including circulation patterns, water temperature, salinity, and turbidity characteristics, also affected the distribution of MPs in the water column [13,18]. A strong correlation was reported between the MPs abundance and the turbidity in the near-bottom water layer [13]. Oceanic fronts were generally formed at the interface of a current system or water with different characteristics (temperature, salinity, or turbidity) [19], which subsequently influenced the MPs occurrence due to the intense dynamics near the frontal zones [20]. The MPs abundance and characteristics in the frontal zones are significant to understanding the MPs pollution at the water-sediment interface. However, until very recently, there was still insufficient work, especially discussing the MPs distribution affected by typical fronts in the bottom water layer.

Owing to the high population density of its catchment and the high consumption of plastics in China, the Yangtze River, ranked among the world's largest rivers, consistently emerges as a primary source of MPs into the ocean in global modeling studies [21,22]. Moreover, the Yangtze River Estuary (YRE) is the gateway where the Yangtze River flows into the East China Sea (ECS), which represents multiple characteristics influenced by interactions between marine and terrestrial environments [21,23]. Due to dense population distribution, river discharge, and various maritime activities, the YRE is vulnerable to plastic accumulation. Consequently, the ecological protection of the YRE and the adjacent area has gained worldwide attention, triggering widespread concern regarding MPs pollution in the YRE and its adjacent sea [18,24–29]. Several studies have reported that the abundance and factors were related to MPs distribution in the surface water or/and sediment of the YRE and ECS [18,24,27,28]. However, to our knowledge, there remains an absence of published works on the MPs distribution and characteristics in the bottom water and the interactions of MPs at the water-sediment interface in the YRE and ECS. Although some other studies reported a high abundance of MPs in the bottom water of other areas [7,8], they still provided limited information about the critical role of the bottom water on the MPs transportation at the water-sediment interface. Thus, the MPs distribution and characteristics at the bottom water in the YRE and ECS, especially the interactions of MPs at the benthic layer, need further investigation and discussion. In addition, there are multi-level fronts outside the mouth of the Yangtze River, which play essential roles in the transport of sediments and the spatial distribution of matters [30], thus greatly affecting the transportation of the sedimentary MPs at the water-sediment interface. Therefore, the relationship between typical fronts and MPs distribution at the water-sediment interface is necessary to be further studied.

Herein, this work selected the YRE and ECS as the study area to reveal the occurrence and distribution of MPs in the benthic layer. In addition to surface water, this study especially explored the water-sediment correlations of MPs, with respect to their occurrence, characteristics, and distribution in the bottom water and surface sediment through network analysis, Principal Co-ordinates Analysis (PCoA), and Pearson correlation analysis. The influences of turbidity on the MPs distribution in the benthic layer were intensively discussed. Moreover, redundancy analysis (RDA) was employed to analyze the correlation between MPs abundance and the environmental parameters. This study will fill the knowledge

gap of the MPs distribution in bottom water and their interactions at the water-sediment interface in the YRE and ECS, thus improving our understanding of the MPs pollution and source-to-sink transport processes.

2. Materials and methods

2.1. Sample collection

Surface water, bottom water, and surface sediment samples were collected at 32 sampling stations in YRE (eight stations) and its adjacent ECS (24 stations) in March 2021 on the cruise of the R/V Zheyuke II (Fig. 1a). The detailed information of the sampling sites and corresponded to the water quality parameters were summarized in Table S1. The sampling sites located at 121°06.912'–123°59.911' E and 29°36.343'–31°45.709' N, with eight sites (B1 to C3) located on both sides of Chongming Island and the other 24 sites (A5-1 to A7-8) in the ECS. The corresponding samples were denoted as SW, BW, and SD, corresponding to those obtained from surface water, bottom water, and surface sediment samples.

The sampling method was selected based on some previous studies [6,13,14] on the investigation of MPs in water and sediment environments and was described briefly below. For each sampling site, 5 L of surface water was collected from the top 20 cm of the water body and filtered through a stainless-steel sieve with a mesh size of 20 μm . 200 mL of bottom water was collected at a 7.50–67.70 m water depth. The water samples were collected through a rosette water sampler in the SBE-911 Plus conductivity-temperature-depth (CTD) system (Sea-Bird Scientific, USA), and environmental variables (temperature, salinity, turbidity, and chlorophyll *a*) were measured simultaneously by sensors on the SBE-911 Plus CTD system. The water samples were filtered through a 20 μm mesh size, and all retained materials on the sieve were repeatedly rinsed into a clean 250 mL blue wide-mouth glass bottle with Milli-Q water. Approximately 1 kg of surface sediment was collected from the top 10 cm layer using a Van Veen grab sampler and then transported into the aluminum foil bags after removing large contaminations or marine organisms. Each sample was taken duplicated at each site. All samples were stored in the dark at 4 °C before analysis.

2.2. Extraction and identification of microplastics

The protocols of MPs extraction from water and sediment followed the procedures of our previous studies [31,32]. Briefly, the water samples were treated with 30% H_2O_2 for 12 h before each was filtered through the GF/C glass microfiber filter paper (47 mm diameter, 0.45 μm pore size), and the sediment samples were separated by flotation. A certain quality of wet sediment (100 g dry weight) was randomly selected to perform with 30% H_2O_2 (v/v). To separate the MPs from the sediment, the samples were floated repeatedly with ZnCl_2 solution (approximately 1.7 g cm^{-3}) by magnetic stirring for 4 h in a beaker. After settling for 48 h, the supernatants were filtered through the filter paper aided by a vacuum pump and then rinsed with Milli-Q water to remove the salt. All the filters after extraction processes were transferred into the glass culture dish and dried at room temperature for further identification and observation. To avoid potential artificial and airborne plastic contamination in the laboratory, all instruments and vessels were carefully rinsed with ultrapure water and tightly wrapped in aluminum foil paper. Cotton laboratory coats and plastic-free gloves were worn during the entire sample collection and laboratory analysis process. Meanwhile, blank experiments were conducted twice to avoid MPs contamination from the

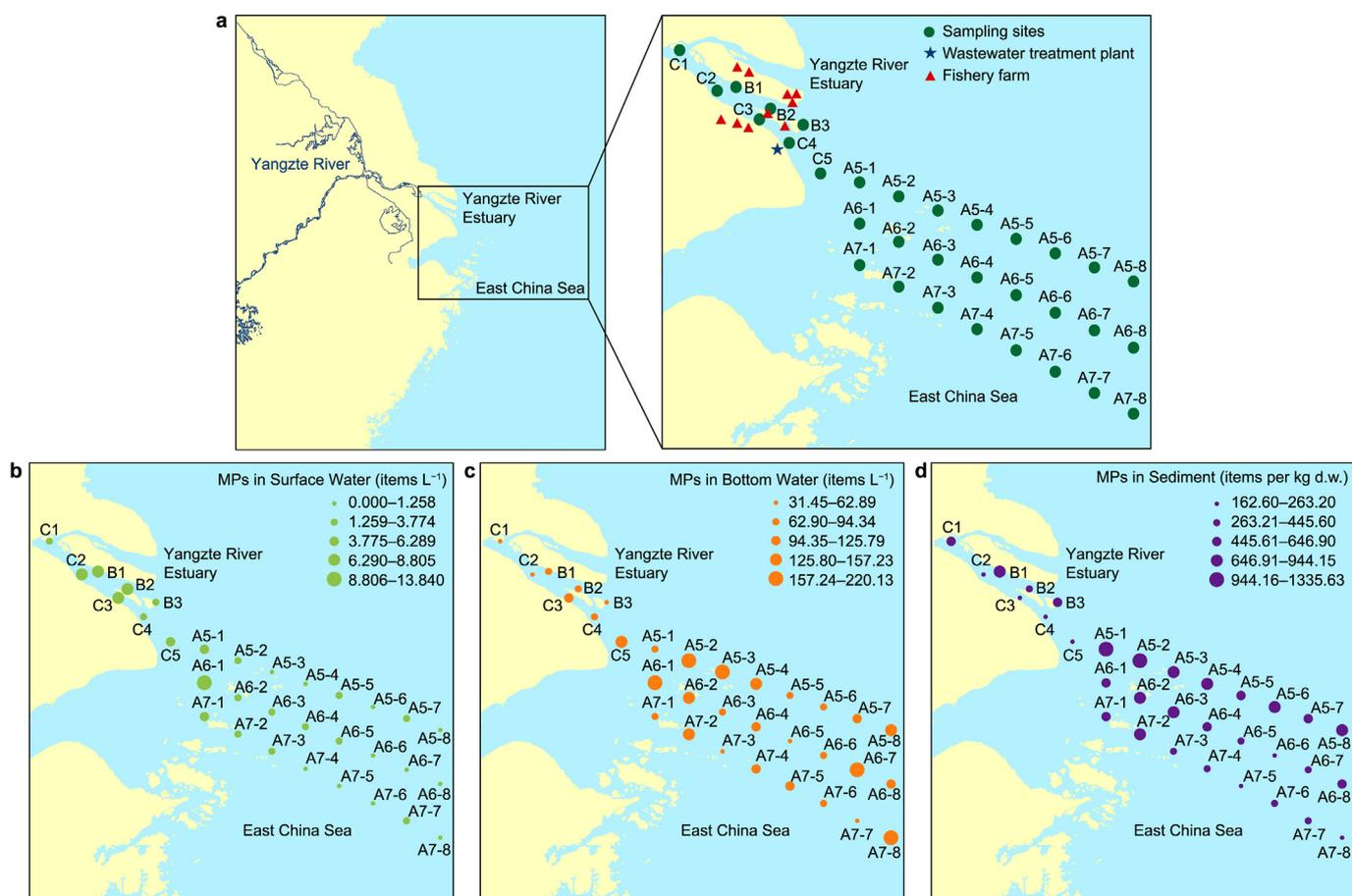


Fig. 1. a, Location of the 32 sampling stations at the YRE and its adjacent Sea (Bailonggang Wastewater Treatment Plant, refer to Refs. [18,27]). b–d, Spatial distribution of MPs abundance in surface water (b), bottom water (c), and sediment (d).

laboratory environment. In addition, the water content of the sediment samples was obtained after oven-drying the wet samples at 60 °C for 48 h.

The particle size of the MPs was sorted into three ranges: 20–100 μm (SPM), 100–500 μm (MPM), and 500–5000 μm (LPM), and the size was measured by Image J (Fiji) software. MPs were assessed visually and categorized by different morphotypes such as sphere, fiber, film, and fragments according to their physical characteristics. The color was classified as transparent/white, black, blue, red, gray, purple, and others (green, pink, yellow, and orange). Moreover, the shape and the color of MPs were recognized by Stereomicroscope (Leica DVM6 V). Polymer types were identified using the micro-Fourier transform infrared spectroscope ($\mu\text{-FTIR}$, Nicolet iN10 MX, Thermo Scientific, USA) equipped with an attenuated total reflection (ATR). Spectra were processed by OMNIC™ Spectra™ software and compared with spectral libraries (including Aldrich Polymers, Hummel Polymer & Additives, Polymer Additives & Plasticizers, Sprouse Polymer Additives, and Synthetic Fibers by Microscope). For accurate polymer identification, particles with spectra matching rates $\geq 70\%$ were accepted. The MPs abundance was calculated by the mapping area of the filter paper via $\mu\text{-FTIR}$. The unit of the MPs abundance in the surface and bottom water was recorded as the number of MPs per liter (items L^{-1}), while that of the sediment was kilogram dry weight (items per kg d.w.).

2.3. Statistical analysis

Graphs of MPs spatial distribution in water and sediment were drawn by ArcGIS 10.8 software (ESRI, CA, USA). Spearman correlation was conducted to display the correlation of water-water or water-sediment systems with respect to the MPs abundance and characteristics using the “magrittr” and “corrplot” packages in R ($R > 0.9$ and $p < 0.05$) [8,33]. Redundancy analysis (RDA) performed via Canoco 5. Pearson correlation was adopted to establish the co-occurrence network of MPs characteristics variables in water and sediment via Gephi-0.9.2 ($R > 0.5$ and $p < 0.05$) (<https://gephi.org/>). The Principal Coordinate Analysis (PCoA) based on Bray-Curtis Distance was performed using PAST 3.0.

3. Results and discussion

3.1. Microplastics abundance and characteristics

The MPs abundance at 32 sampling sites in the surface water, bottom water, and sediment of the YRE and ECS was illustrated in Fig. 2 with detailed data in Table S2. The MPs abundance ranged from 1.26 to 13.84 items L^{-1} with an average of 3.69 ± 2.87 items L^{-1} (surface water), 31.45 to 220.14 items L^{-1} with an average of 118.91 ± 46.39 items L^{-1} (bottom water), and 16.00 to 1335.00 items per kg d.w. with an average of 544.15 ± 305.70 items per kg d.w. (sediment), respectively. The MPs abundance in the YRE and

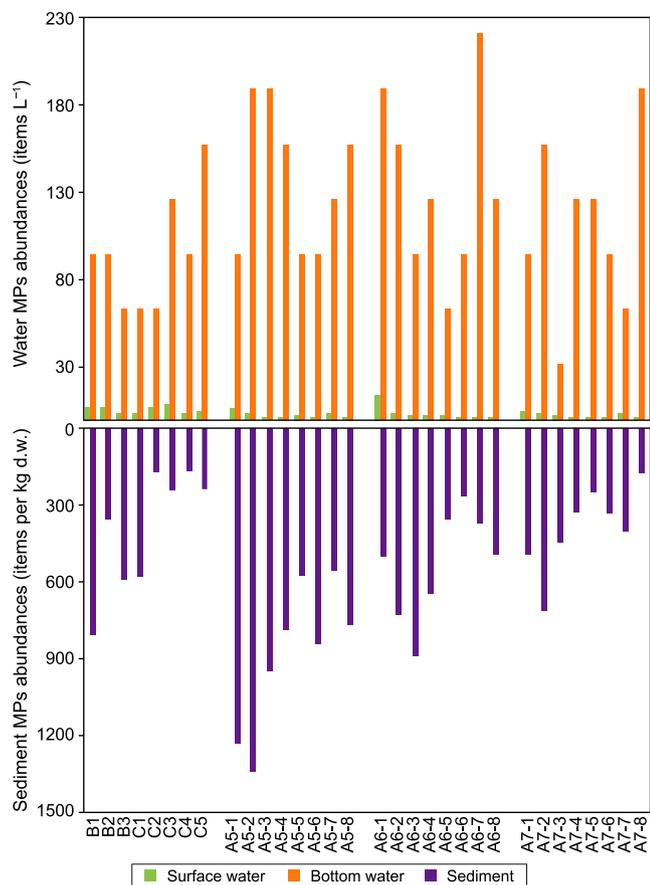


Fig. 2. The abundance of MPs in the surface water, bottom water, and sediment of 32 sampling sites in YRE and ECS.

Table 1 Comparison of MPs abundance in water and sediment from different watershed estuaries.

Watershed	System	Abundance range ^a	Mean abundance ^a	Sampling time	Sampling methods	Reference
Yangtze River Estuary & East China Sea	Surface water	1.26–13.84 items L ⁻¹	3.69 ± 2.87 items L ⁻¹	March 2020	20 μm mesh	This study
Yangtze River Estuary	Surface water	0.50–10.20 items L ⁻¹	4.14 ± 2.46 items L ⁻¹	July 2013	333 μm mesh	[24]
Pearl River Estuary	Surface water	7.85–10.95 items L ⁻¹	8.90 items L ⁻¹	December 2017	50 μm mesh	[35]
Bohai Bay (Haihe Estuary)	Surface water	-	1.49 ± 0.82 items L ⁻¹	-	-	[26]
Bohai Bay (Yongdingxinhe Estuary)	-	-	7.88 ± 0.46 items L ⁻¹	-	-	[28]
Yangtze River Estuary (Chongming Island)	Surface water	0–0.26 items L ⁻¹	-	June 2019	300 μm mesh	[28]
Lower Yellow River near Estuary	Surface water	380.00–582.00 items L ⁻¹ (wet seasons)	-	July 2018	50 μm mesh	[34]
		623.00–1392.00 items L ⁻¹ (dry seasons)	-	March 2019	50 μm mesh	[34]
Yangtze River Estuary & East China Sea	Bottom water	31.45–220.14 items L ⁻¹	118.91 ± 46.39 items L ⁻¹	March 2020	20 μm mesh	This study
Yangtze River Estuary & East China Sea	Sediment	162.00–1335.00 items per kg d.w.	544.15 ± 305.70 items per kg d.w.	March 2020	0–10 cm	This study
Yangtze River Estuary	Sediment	20.00–340.00 items per kg d.w.	121.00 ± 9.00 items per kg d.w.	September 2015	5–10 cm	[27]
Bohai Bay (Haihe Estuary)	Sediment	-	216.10 ± 92.10 items per kg d.w.	-	-	[26]
Bohai Bay (Yongdingxinhe Estuary)	Sediment	-	85.0 ± 40.10 items per kg d.w.	-	-	[26]
Yellow Sea and East China Sea	Sediment	60.00–240.00 items per kg d.w.	-	March 2017	Upper 10 cm	[25]
Pearl River Estuary	Sediment	100.00–7900.00 items per kg d.w.	851.00 ± 177.00 items per kg d.w.	November 2015	Top 5 cm	[37]

^a The unit of MPs abundance in water was normalized as “items L⁻¹” for convenient comparison.

ECS of this study was compared with the data published in other works in this area and other major estuaries in China (Table 1) [26,28,34–37]. As shown in Table 1, the MPs abundance in surface water of this study is less abundant compared with that reported in the Pearl River Estuary [35] and Bohai Bay (such as Yongding New Estuary) [26], but a moderate level value was found for the sediment. In comparison with those previously reported data in YRE and ECS by other studies [24,36], the data obtained in this study showed a comparable level for the surface water but a higher average value in the sediment, which may be caused by the variances in the sampling methodology like different mesh sieves for filtering, different sampling time (year and season) and sites [18]. Most importantly, the MPs in the bottom water of the YRE and ECS were recorded for the first time in this study, showing the noticeably higher MPs abundance in the bottom water than those in the surface water.

MPs characteristics, including particle size, shape, color, and polymer types of all samples collected from the surface water, bottom water, and sediment, are summarized in Fig. 3a–d. Large particle MPs with the size range of 500–5000 μm (LPM) were dominant at all sampling sites, comprising 64.0 ± 24.0% of the total particle number (Fig. 3a). Specifically, fibers were predominant (7.0 ± 11.0%) together with a much smaller amount of fragments (10.0 ± 13.0%) and films (10.0 ± 13.0%) (Fig. 3b). The predominance of fibers was derived from wastewater discharge by clothes washing and textile industries [38,39], and the maritime activities (e.g., packaging, fishing gear) [40,41]. As shown in Fig. 3c, a wide array of colors was detected for MPs at all sampling sites, while the colored MPs (including blue, red, purple, green, pink, etc.) comprised the majority of MPs at all sampling locations, which were commonly used in packaging, clothing materials, and many other applications [18]. Among the colorful MPs, blue MPs were the most abundant (47.0 ± 26.0%). Several factors may contribute to this prevalence. Firstly, blue is a universally prevalent color (jeans, shirts, etc.) and

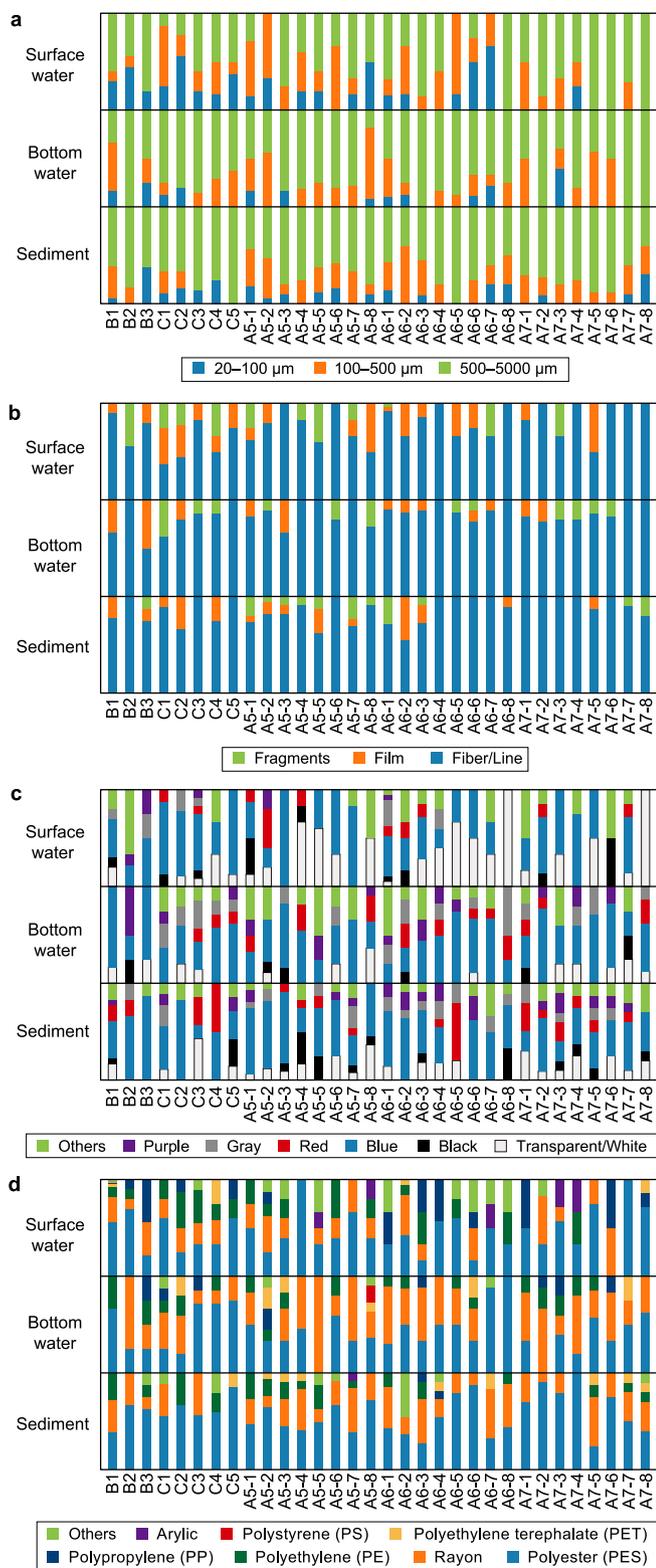


Fig. 3. The characteristic distribution of MPs in surface water, bottom water, and sediment of 32 sampling sites, including size (a), shape (b), color (c), and polymer type (d).

may not be attractive for ingestion [42]. Secondly, fishing nets and ropes, widely used in the fishing industry, may represent additional sources of blue MPs [43]. Furthermore, decolorized microplastics

were discovered at various locations as the original color of these microplastics had faded due to weathering processes [27]. Notably, the MPs extraction process in this study could not influence the MPs color and transparency. In terms of polymer types (Fig. 3d), polyester (PES), rayon, polyethylene (PE), polypropylene (PP), polyethylene terephthalate (PET), acrylic, polystyrene (PS), others (Nylon, PP-PE copolymer, etc.) were identified. Fig. S1 presents the representative polymer types and their corresponding microscope images. The PES (49.0 ± 22.0%) and rayon (27.0 ± 20.0%), as the component of synthetic fibers, were dominantly found in the sampling sites. Besides, other common polymer types were also found, such as PE (12.0 ± 13.0%) and PP (2.0 ± 6.0%) as the widely used plastic types [44]. The PES and rayon in both water and sediment might be released from clothes washing and textile industries [38,39], while the sources of PE and PP can be derived from packaging, containers, pipes, agricultural film, automotive parts, fishing gears, and houseware [44].

The network analysis was conducted with particle size, shape, color, and type of MPs in water and sediment based on Pearson's correlation analysis to reveal the co-occurrence correlations among MPs characteristics (Fig. S2). The network analysis showed that fibrous MPs and films mainly were PES with a large size (LPM) and PP as transparent/white. PE mainly existed as films and fragments with a blue color in a small size range (SPM). In addition, a positive correlation was observed between the SPM as the size and the film/fragments as the shape. This indicated that films and fragments were present in the water and sediment with small sizes.

3.2. Horizontal and vertical distribution characteristics of MPs

The spatial (horizontal and vertical) distribution of MPs abundance in the surface water, bottom water, and sediment in all sampling sites are illustrated in Fig. 1b–d, showing their wide existence and distinct variation (indicated by circle sizes). For the surface water, more MPs were mainly observed in the estuary and the saltwater-freshwater interface, indicating decreased MPs abundance with prolonged distance from the shore. For the sediment, MPs abundance was comparatively higher in the inshore areas than in estuary and far shore areas. Notably, cross-section A5 defied this trend by displaying higher MPs abundance in all sampling sites of this section. Similarly, relatively more abundant MPs were also detected in the bottom water of the inshore area, but without an obvious trend from nearshore to farther sites. However, several far shore sites of cross-sections A5–A7 were also observed with higher MPs abundance. Among all sampling sites, the highest MPs abundance was recorded as 13.84 items L⁻¹ of the surface water at site A6-1, 1335.63 items per kg d.w. of the sediment at site A5-2, and 220.14 items L⁻¹ of the bottom water at site A6-7. For the YRE, the ocean is well stratified with light, fresh water on the surface and dense salty water in the bottom layer. The pycnocline separates the water column into two parts with different densities. Considering buoyancy, the low-density MPs will stay in the surface layer (freshwater), while the high-density MPs will sink into the bottom layer (salt water). As seawater density is mainly decided by the temperature and salinity, we found significant correlations between MPs abundance and temperature and salinity for surface water in Fig. S3. However, the real ocean is more complicated; dynamic processes will also affect the distribution of MPs in the seawater. Along the pycnocline, friction caused by the horizontal flow will induce vertical mixing and raise the denser particles; strong tidal flow will even resuspend the particles up (sediments, including MPs). Horizontal ocean currents could also transport MPs, leading to different horizontal distributions. Furthermore, ocean fronts will be a barrier limiting horizontal transportation. All these physical processes could lead to control of both vertical and

horizontal distributions of MPs.

Furthermore, Fig. 3 illustrates the variation of MPs characteristics among the sampling sites in the YRE (eight sites) and ECS (24 sites). Higher amounts of SPM (20–100 μm) were observed in the estuary, but MPs with larger sizes were found in the sea areas, especially in the sampling sites of cross-section A7. Although fibers were the predominant MPs shape, the higher variation in MPs shape was investigated in the surface water of the estuary. No apparent tendency was detected for the MPs color and polymer type among the sampling sites in different areas. From the perspective of vertical distribution, a much higher MPs amount was observed in the bottom water than in the surface water (Fig. 2), indicating the significant variation of MPs distribution in waters affected by depth [45].

The vertical distribution of MPs characteristics is further depicted in Fig. 4, showing the dominance of LPM (size, 52.4–66.7%), fibers (shape, 76.8–86.2%), blue (color, 46.0–47.9%), and PES and rayon (polymer types, 64.7–82.8%) in the water to sediment samples. Besides, the surface water exhibited the highest ratio of minor species with respect to each characteristic of the MPs, in comparison with the corresponding value of bottom water and sediment. For example, with lower densities [9,35], more PE and PP were found in the surface water (23.0%) than the bottom water (13.0%) and sediment (10.1%), while transparent/white MPs were more likely to exist in the surface water (20.5%) other than those in the sediment (13.0%) and bottom water (7.2%).

The spatial redistribution of MPs was affected by point source discharges, biological activities, and vertical/horizontal transport of the MPs [9]. As demonstrated in Fig. 1a, some wastewater treatment plants and fishery farms located in the river estuary caused

higher MPs abundance directly in the surface water of this area, as reflected in Fig. 2. In this area, Bailonggang Wastewater Treatment Plant is one of the largest sewage treatment plants, with a capacity of 1.2 million m³ d⁻¹ and accounting for almost 25% of the total capacity of wastewater treatment in Shanghai, China [46]. Although 90% of MPs can be removed after treatment [47], the remaining plastics can still be transported from land to sea. After MPs are released, the monsoons, currents, Changjiang plume, and tides may further affect the MPs distribution [48]. The strong hydrological force of the estuary carried plastic waste from the land to the sea, resulting in higher MPs abundances in the bottom water and sediment of the inshore area because of MPs deposition [18,21,23,28]. Moreover, the Changjiang diluted water and other water masses moved from the mouth of the estuary section C to A5, which brought MPs particles from the estuary to the northeastern region [18]. Therefore, being carried by coastal circulation, more MPs were subsequently deposited and accumulated in the sediment of the northeast part of the study area. In addition, the similar MPs characteristics between the eight sites in the estuary and 24 sites of the sea (Fig. 3) further indicated the importance of terrestrial MPs sources from the river to its adjacent sea. Theoretically, MPs with high densities are more likely to deposit while those with low densities are prone to be transported over a relatively long distance [2], which is consistent with our results that higher proportions of PES+Rayon+PET in the sediment and more PE+PP in the surface water. However, the vertical distribution of MPs at the water-sediment interface was also affected by multifactorial hydrodynamics (Ekman pumping, upwelling, and downwelling, etc.) [49,50], which could explain the majority of PES+Rayon+PET in the bottom water.

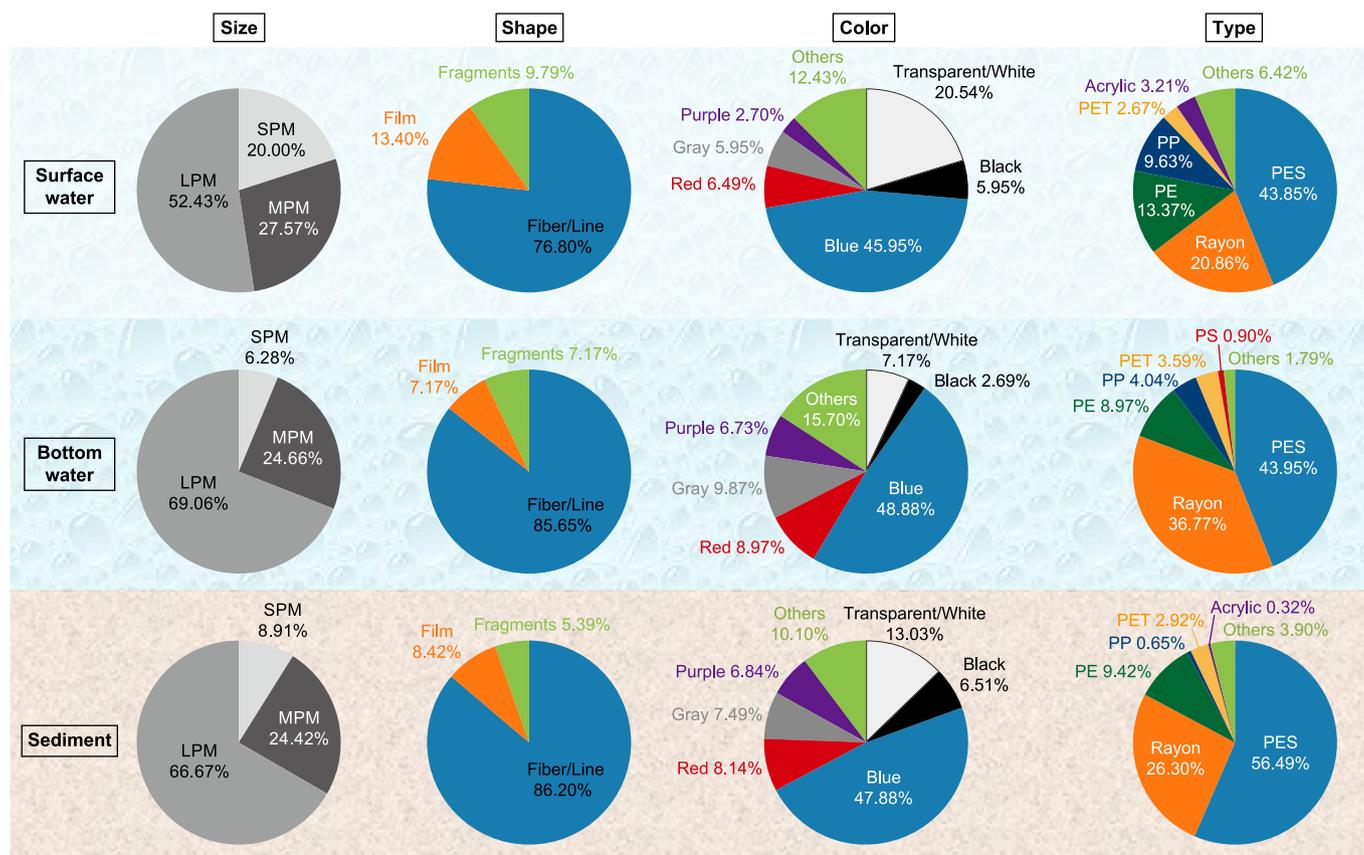


Fig. 4. Vertical distribution of the MPs among surface water, bottom water, and sediment in different characteristics (size, shape, color, and polymer type).

3.3. Microplastics in the benthic layer

3.3.1. Interactions of microplastics in water and sediment

To reveal the co-occurrence correlations of MPs among the surface water, bottom water, and sediment, a network based on Spearman correlation analysis (Fig. 5a) was applied with respect to the data of MPs abundance, particle size, shape, color, and polymer types. In the bottom water, a strongly positive correlation was found for C5 vs. A6-4 and A5-4 vs. A5-7, while in the sediment, the sites were A5-6 vs. A7-2, A5-8 vs. A7-8. In the surface water, the highly correlated sites were A5-6 vs. A7-1 and A6-8 vs. A7-8. The correlation results among the sampling sites suggested that the horizontal migration of the MPs could cause a close relationship between nearby locations. Furthermore, significant correlations were also observed between some sites in the vertical direction, especially between the bottom water and sediment. For example, the MPs at SD_B2 were highly correlated with those at BW_C5 and BW_A6-4, respectively. Meanwhile, the following sites were also highly interacted, e.g., SD_A7-3 vs. BW_C5, SD_A6-1 vs. BW_A7-4, and SD_C2 vs. BW_B1. It indicated that the MPs in the bottom water and the sediment were more strongly related than the surface water. This further suggested that the bottom water might mediate the MPs distribution at water-sediment. The critical role of the bottom water layer for the vertical distribution of other pollutants was also reported, showing the highest level of di(2-ethylhexyl) phthalate (DEHP) in the bottom layer of the water

column and a significant correlation between the bottom water layer and the bed sediment [51]. Thus, the resuspension and deposition of MPs at the water-sediment system led to the much higher MPs abundance in the bottom water.

The PCoA (Fig. 5b) showed a much stronger correlation of the variables between the sediment and bottom water, consistent with the network analysis results in Fig. 5a, further proving the critical role of bottom water on MPs distribution at the benthic layer. Besides, as shown in Fig. 5b, the diversity of MPs among different sampling sites was the highest in the bottom water, followed by the sediment and the surface water. The complex distribution of MPs in the bottom water can be explained by the reasons that selective sedimentation of some high-density MPs and MPs resuspension from sediment to bottom water caused by disturbance [16,17,52]. Moreover, the redundancy analysis (Fig. S4a) was applied to reflect the relationship of MPs types between the bottom water and sediment. For instance, Rayon, PES, and PE in the bottom water were primarily affected by the MPs type in the sediment. To specify, a positive correlation was found for the PE in the bottom water and the sediment, while a negative correlation was observed for the Rayon and PES. The MPs with high density (e.g., Rayon and PES) tended to deposit in the sediment, while the low-density MPs (e.g., PE) were prone to transport over a relatively longer distance [45]. Therefore, these results demonstrated that the bottom water mediated the distribution and transportation of MPs at water-sediment.

3.3.2. Impact of environmental factors on MPs distribution in the benthic layer

Pearson's correlation analysis was conducted among the MPs abundance (surface and bottom water), the longitude and latitude of the sampling sites, sampling water depth, as well as the water temperature, salinity, content of chlorophyll *a*, and turbidity, with results illustrated in Fig. S3. According to the correlation analysis, an obvious correlation in the surface water was found between the MPs abundance and the environmental parameters above (except the sampling water depth). However, there was no significant correlation between the MPs abundance in the bottom water and these parameters. This might be because the distribution of MPs in the bottom water was more likely to be affected by sediment disturbance. In addition, there was a significant correlation ($p < 0.001$) between the MPs abundance and temperature in the surface water (Fig. S3). The density of the seawater in the semi-enclosed sea varies considerably with temperature and salinity, which potentially causes changes in MPs transportation when the salinity or temperature changes [53]. The high flow velocity in the bottom water layer was observed to facilitate the transport of sedimental MPs [45]. The MPs in the surface sediment could transfer to both overlying water and deeper sediment during the disturbance process [54]. Iribarne et al. [52] also reported that the disturbance processes, such as trawling and bioturbation, influenced the distribution of plastic litter in marine sediment and the overlying water column. In addition, dynamic processes of the seawater will also control both the horizontal and vertical distribution of MPs, including the turbulence mixing-induced vertical motion and resuspension in the bottom, ocean currents-induced horizontal transportation, and convergence along the ocean fronts.

Rather than the general environmental parameters, the disturbance-induced resuspension process may significantly affect MPs' distribution and migration in the benthic layer. As shown in Fig. S4b, a positive correlation was observed between MPs abundance in the sediment and the turbidity of the bottom water, which suggested that either the resuspension of surface sediment or the sedimentation could influence the turbidity of the bottom water, further affecting the MPs distribution in the benthic layer. Another

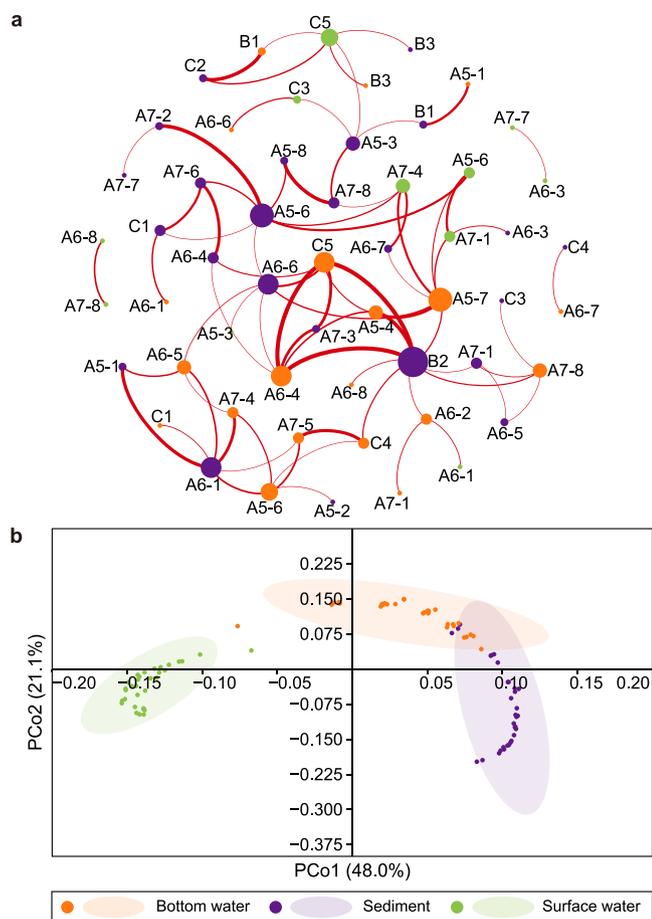


Fig. 5. a, Co-occurrence analysis based on the Spearman correlation analysis using network ($R > 0.9$, $p < 0.05$). b, PCoA based on the Bray-Curtis Distance for MPs variables (abundance and characteristics) in the surface water, bottom water, and sediment of the sampling sites.

recent study also reported that bottom currents could firmly control the movement of MPs at the seafloor and regulate their vertical distribution [55]. Thus, a further analysis was conducted based on the interactions for sections A5, A6, and A7. Beyond the sites with the maximum turbidity (A5-1, A6-1, A7-1), the MPs abundance of the bottom water declined accordingly with the turbidity decreasing (A5-2 to A5-5, A6-2 to A6-5, and A7-4 to A7-8). This phenomenon can be explained by the effects of the upwelling in the winter (cold currents) in the research region of the ECS [56]. The strongest upwelling region tends to appear in the turbidity front zone, which may also be related to the resuspension of the sediment. Upwelling was probably an efficient pathway for the transport of MPs from the bottom water to surface water [9]. It could be concluded that the gradually decreased MPs abundance at those sampling sites might be due to the resuspension process of bottom water through upwelling. Therefore, the resuspension processes largely affected the distribution of MPs in the benthic layer.

4. Conclusions and perspectives

This study comprehensively investigated the occurrence of horizontal and vertical distribution characteristics of MPs in the YRE and ECS. A particular emphasis is placed on understanding the critical role of the benthic layer in MPs distribution and transportation. Notably, we discovered a significant abundance of MPs in the bottom water for the first time in this region, surpassing the quantity found in surface water by 8–175 times. Also, higher amounts of MPs in the sediment were obtained in this research area compared to the previous studies. The similar MPs characteristics distribution in the bottom water with sediment, 0.5–5.0 mm particle size, fibrous and colorful MPs particles, as well as PES and rayon, accounted for a larger percentage at most sampling sites. Besides, large amounts of blue MPs were observed in the research area. These morphological characteristics, density, and chemical compositions of MPs provided clues about their potential sources of MPs pollution in the region, primarily originating from terrestrial discharge and fishery activities. The co-occurrence network analysis results showed strong correlations among some sites owing to the horizontal transportation of MPs, as well as the two compartments of bottom water and sediment due to the influence of resuspension and deposition processes. In addition, PCoA further revealed the strong correlation between the bottom water and sediment in the benthic layer, indicating the critical role of the bottom water as a mediator in the vertical distribution of MPs. Moreover, a positive correlation was also found for the MPs abundance in the sediment and the turbidity of the bottom water, highlighting the predominant influence of resuspension processes on MPs distribution in the benthic layer. Overall, this study mainly focused on a field investigation and preliminary discussion to indicate the critical role of the benthic layer in the vertical distribution of MPs. However, the natural environment is inherently complex, making it challenging to precisely elucidate the multifaceted impacts of various factors, such as temperature, salinity, and water turbidity, via field investigation. Therefore, future laboratory experiments should be intensively conducted to deeply explore the details of multiple environmental factors affecting the distribution and transportation of MPs in the benthic layer.

Credit authorship contribution statement

Xinyu Ge: Investigation, Data Curation, Visualization, Writing - Original Draft. **Feng Xu:** Data Curation, Visualization, Writing - Original Draft. **Yuanyuan Tang & Lili Liu & Bo Li:** Conceptualization, Resources, Funding Acquisition, Project Administration, Writing - Review & Editing. **Xiao Lu & Lijuan Wang:** Investigation, Data Curation. **Jiangpeng Li:** Methodology.

Data availability

Data will be made available on request.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.ese.2023.100340>.

References

- [1] M. Cole, P. Lindeque, C. Halsband, T.S. Galloway, Microplastics as contaminants in the marine environment: a review, *Mar. Pollut. Bull.* 62 (2011) 2588–2597.
- [2] J. Fan, L. Zou, T. Duan, L. Qin, Z. Qi, J. Sun, Occurrence and distribution of microplastics in surface water and sediments in China's inland water systems: a critical review, *J. Clean. Prod.* 331 (2022) 129968–129981.
- [3] L.C. De Sá, M. Oliveira, F. Ribeiro, T.L. Rocha, M.N. Futter, Studies of the effects of microplastics on aquatic organisms: what do we know and where should we focus our efforts in the future? *Sci. Total Environ.* 645 (2018) 1029–1039.
- [4] W. Wang, H. Gao, S. Jin, R. Li, G. Na, The ecotoxicological effects of microplastics on aquatic food web, from primary producer to human: a review, *Ecotoxicol. Environ. Saf.* 173 (2019) 110–117.
- [5] Y. Zhang, S. Kang, S. Allen, D. Allen, T. Gao, M. Sillanpää, Atmospheric microplastics: a review on current status and perspectives, *Earth Sci. Rev.* 203 (2020) 103118–103133.
- [6] P.L. Lenaker, A.K. Baldwin, S.R. Corsi, S.A. Mason, P.C. Reneau, J.W. Scott, Vertical distribution of microplastics in the water column and surficial sediment from the Milwaukee River basin to lake Michigan, *Environ. Sci. Technol.* 53 (2019) 12227–12237.
- [7] A.R.A. Lima, M.F. Costa, M. Barletta, Distribution patterns of microplastics within the plankton of a tropical estuary, *Environ. Res.* 132 (2014) 146–155.
- [8] L. Liu, F. Sun, H. Zhao, H. Mi, S. He, Y. Chen, Y. Liu, H. Lan, M. Zhang, Z. Wang, Compositional changes of sedimentary microbes in the Yangtze River Estuary and their roles in the biochemical cycle, *Sci. Total Environ.* 760 (2021) 143383–143398.
- [9] X. Wang, L. Zhu, K. Liu, D. Li, Prevalence of microplastic fibers in the marginal sea water column off southeast China, *Sci. Total Environ.* 804 (2022) 150138–150146.
- [10] J. Martin, A. Lusher, R.C. Thompson, A. Morley, The deposition and accumulation of microplastics in marine sediments and bottom water from the Irish continental shelf, *Sci. Rep.* 7 (2017) 1–9.
- [11] L. Van Cauwenberghe, L. Devriese, F. Galgani, J. Robbins, C.R. Janssen, Microplastics in sediments: a review of techniques, occurrence and effects, *Mar. Environ. Res.* 111 (2015) 5–17.
- [12] P. Yao, B. Zhou, Y.H. Lu, Y. Yin, Y.Q. Zong, M.T. Chen, Z. O'Donnell, A review of microplastics in sediments: spatial and temporal occurrences, biological effects, and analytic methods, *Quat. Int.* 519 (2019) 274–281.
- [13] Q. Zhou, C. Tu, J. Yang, C. Fu, Y. Li, J.J. Waniek, Trapping of microplastics in halocline and turbidity layers of the semi-enclosed baltic sea, *Front. Mar. Sci.* 8 (2021) 1–13.
- [14] Y.K. Song, S.H. Hong, S. Eo, M. Jang, G.M. Han, A. Isobe, W.J. Shim, Horizontal and vertical distribution of microplastics in Korean coastal waters, *Environ. Sci. Technol.* 52 (2018) 12188–12197.
- [15] R.S. Pazos, D.E. Bauer, N. Gómez, Microplastics integrating the coastal planktonic community in the inner zone of the Río de la Plata estuary (South America), *Environ. Pollut.* 243 (2018) 134–142.
- [16] M. Kooi, J. Reisser, B. Slat, F.F. Ferrari, M.S. Schmid, S. Cunsolo, R. Brambini, K. Noble, L.A. Sirks, T.E.W. Linders, R.I. Schoeneich-Argent, A.A. Koelmans, The effect of particle properties on the depth profile of buoyant plastics in the ocean, *Sci. Rep.* 6 (2016) 1–10.
- [17] K. Liu, W. Courtene-Jones, X. Wang, Z. Song, N. Wei, D. Li, Elucidating the vertical transport of microplastics in the water column: a review of sampling

- methodologies and distributions, *Water Res.* 186 (2020), 116403–116215.
- [18] P. Xu, G. Peng, L. Su, Y. Gao, L. Gao, D. Li, Microplastic risk assessment in surface waters: a case study in the Changjiang Estuary, China, *Mar. Pollut. Bull.* 133 (2018) 647–654.
- [19] I.M. Belkin, P.C. Cornillon, K. Sherman, Fronts in large marine ecosystems, *Prog. Oceanogr.* 81 (2009) 223–236.
- [20] C. Lu, P.W. Kania, K. Buchmann, Particle effects on fish gills: an immunogenetic approach for rainbow trout and zebrafish, *Aquaculture* 484 (2018) 98–104.
- [21] L.C.M. Lebreton, J. Van Der Zwet, J.W. Damsteeg, B. Slat, A. Andrady, J. Reisser, River plastic emissions to the world's oceans, *Nat. Commun.* 8 (2017) 1–10.
- [22] X. Xiong, C. Wu, J.J. Elser, Z. Mei, Y. Hao, Occurrence and fate of microplastic debris in middle and lower reaches of the Yangtze River – from inland to the sea, *Sci. Total Environ.* 659 (2019) 66–73.
- [23] S. Zhao, L. Zhu, T. Wang, D. Li, Suspended microplastics in the surface water of the Yangtze Estuary System, China: first observations on occurrence, distribution, *Mar. Pollut. Bull.* 86 (2014) 562–568.
- [24] S. Zhao, T. Wang, L. Zhu, P. Xu, X. Wang, L. Gao, D. Li, Analysis of suspended microplastics in the Changjiang Estuary: implications for riverine plastic load to the ocean, *Water Res.* 161 (2019) 560–569.
- [25] F. Zhang, X. Wang, J. Xu, L. Zhu, G. Peng, P. Xu, D. Li, Food-web transfer of microplastics between wild caught fish and crustaceans in East China Sea, *Mar. Pollut. Bull.* 146 (2019) 173–182.
- [26] N. Wu, Y. Zhang, X. Zhang, Z. Zhao, J. He, W. Li, Y. Ma, Z. Niu, Occurrence and distribution of microplastics in the surface water and sediment of two typical estuaries in Bohai Bay, China, *Environ. Sci. Process. Impacts.* 21 (2019) 1143–1152.
- [27] G. Peng, B. Zhu, D. Yang, L. Su, H. Shi, D. Li, Microplastics in sediments of the Changjiang estuary, China, *Environ. Pollut.* 225 (2017) 283–290.
- [28] Y. Li, Z. Lu, H. Zheng, J. Wang, C. Chen, Microplastics in surface water and sediments of chongming Island in the Yangtze estuary, China, *Environ. Sci. Eur.* 32 (2020) 1–12.
- [29] J. Li, B. Hu, H. Wei, J. Zhao, L. Zou, F. Bai, Y. Dou, L. Wang, X. Fang, Provenance variations in the Holocene deposits from the southern Yellow Sea: clay mineralogy evidence, *Continental Shelf Res.* 90 (2014) 41–51.
- [30] L. Ye, K. Taguchi, S. Zhang, Characterization of hydrodynamic and sediment transport processes in the East China Sea, *J. Adv. Mar. Sci. Technol. Soc.* 18 (2012) 27–62.
- [31] P. Wu, Y. Tang, M. Dang, S. Wang, H. Jin, Y. Liu, H. Jing, C. Zheng, S. Yi, Z. Cai, Spatial-temporal distribution of microplastics in surface water and sediments of maozhou river within Guangdong-Hong Kong-Macao greater Bay area, *Sci. Total Environ.* 717 (2020) 135187–135198.
- [32] L. Chen, J. Li, Y. Tang, S. Wang, X. Lu, Z. Cheng, X. Zhang, P. Wu, X. Chang, Y. Xia, Typhoon-induced turbulence redistributed microplastics in coastal areas and reformed plastisphere community, *Water Res.* 204 (2021) 117580–117592.
- [33] W. Wan, J. Tan, Y. Wang, Y. Qin, H. He, H. Wu, W. Zuo, D. He, Responses of the rhizosphere bacterial community in acidic crop soil to pH: changes in diversity, composition, interaction, and function, *Sci. Total Environ.* 700 (2020) 134418–134428.
- [34] M. Han, X. Niu, M. Tang, B.T. Zhang, G. Wang, W. Yue, X. Kong, J. Zhu, Distribution of microplastics in surface water of the lower Yellow River near estuary, *Sci. Total Environ.* 707 (2020) 135601–135610.
- [35] M. Yan, H. Nie, K. Xu, Y. He, Y. Hu, Y. Huang, J. Wang, Microplastic abundance, distribution and composition in the Pearl River along guangzhou city and Pearl River estuary, China, *Chemosphere* 217 (2019) 879–886.
- [36] C. Zhang, H. Zhou, Y. Cui, C. Wang, Y. Li, D. Zhang, Microplastics in offshore sediment in the Yellow Sea and East China Sea, China, *Environ. Pollut.* 244 (2019) 827–833.
- [37] L. Zuo, Y. Sun, H. Li, Y. Hu, L. Lin, J. Peng, X. Xu, Microplastics in mangrove sediments of the Pearl River Estuary, South China: correlation with halogenated flame retardants' levels, *Sci. Total Environ.* 725 (2020) 138344–138351.
- [38] M.A. Browne, P. Crump, S.J. Niven, E. Teuten, A. Tonkin, T. Galloway, R. Thompson, Accumulation of microplastic on shorelines worldwide: sources and sinks, *Environ. Sci. Technol.* 45 (2011) 9175–9179.
- [39] S. Wei, H. Luo, J. Zou, J. Chen, X. Pan, D.P.L. Rousseau, J. Li, Characteristics and removal of microplastics in rural domestic wastewater treatment facilities of China, *Sci. Total Environ.* 739 (2020), 139935–129944.
- [40] N. Soltani, B. Keshavarzi, F. Moore, R. Busquets, M.J. Nematollahi, R. Javid, S. Gobert, Effect of land use on microplastic pollution in a major boundary waterway: the Arvand River, *Sci. Total Environ.* 830 (2022) 154728–154743.
- [41] B. Xue, L. Zhang, R. Li, Y. Wang, J. Guo, K. Yu, S. Wang, Underestimated microplastic pollution derived from fishery activities and “hidden” in deep sediment, *Environ. Sci. Technol.* 54 (2020) 2210–2217.
- [42] J. Gago, O. Carretero, A.V. Filgueiras, L. Viñas, Synthetic microfibers in the marine environment: a review on their occurrence in seawater and sediments, *Mar. Pollut. Bull.* 127 (2018) 365–376.
- [43] D.V. Dantas, M. Barletta, M.F. da Costa, The seasonal and spatial patterns of ingestion of polyfilament nylon fragments by estuarine drums (Sciaenidae), *Environ. Sci. Pollut. Res.* 19 (2012) 600–606.
- [44] Plastics Europe, *Plastics-the Facts 2020, an Analysis of European Plastics Production, Demand and Waste Data*, 2020.
- [45] B. He, M. Smith, P. Egodawatta, G.A. Ayoko, L. Rintoul, A. Goonetilleke, Dispersal and transport of microplastics in river sediments, *Environ. Pollut.* 279 (2021) 116884–116893.
- [46] L.H. Wang, X.Q. Zha, Q. Shao, Design and commissioning of sludge anaerobic digestion system in Bailonggang wastewater treatment plant, *China Water & Wastewater* 28 (2012) 43–48.
- [47] J. Talvitie, A. Mikola, A. Koistinen, O. Setälä, Solutions to microplastic pollution—removal of microplastics from wastewater effluent with advanced wastewater treatment technologies, *Water Res.* 123 (2017) 401–407.
- [48] S. Li, H. Wang, D. Liang, Y. Li, Z. Shen, How the Yangtze River transports microplastic to the east China sea, *Chemosphere* 307 (2022) 136112–136121.
- [49] E. Van Sebille, S. Aliani, K.L. Law, N. Maximenko, J.M. Alsina, A. Bagaev, D. Wichmann, The physical oceanography of the transport of floating marine debris, *Environ. Res. Lett.* 15 (2020) 23003–23036.
- [50] H.J. Cunningham, C. Higgins, T.S. van den Bremer, The role of the unsteady surface wave-driven Ekman–Stokes flow in the accumulation of floating marine litter, *J. Geophys. Res. Ocean.* 127 (2022) 1–19.
- [51] A. Paluselli, S.K. Kim, Horizontal and vertical distribution of phthalates acid ester (PAEs) in seawater and sediment of East China Sea and Korean South Sea: traces of plastic debris? *Mar. Pollut. Bull.* 151 (2020) 110831–110840.
- [52] O. Iribarne, F. Botto, P. Martinetto, J.L. Gutierrez, The role of burrows of the SW Atlantic intertidal crab *Chasmagnathus granulata* in trapping debris, *Mar. Pollut. Bull.* 40 (2000) 1057–1062.
- [53] A. Bagaev, A. Mizyuk, L. Khatmullina, I. Isachenko, I. Chubarenko, Anthropogenic fibres in the Baltic sea water column: field data, laboratory and numerical testing of their motion, *Sci. Total Environ.* 599 (2017) 560–571.
- [54] F. Xia, Q. Yao, J. Zhang, D. Wang, Effects of seasonal variation and resuspension on microplastics in river sediments, *Environ. Pollut.* 286 (2021) 117403–117413.
- [55] B. Zhang, L. Chen, J. Chao, X. Yang, Q. Wang, Research progress of microplastics in freshwater sediments in China, *Environ. Sci. Pollut. Res.* 27 (2020) 31046–31060.
- [56] F. Qiao, Y. Yang, X. Lu, C. Xia, X. Chen, B. Wang, Y. Yuan, Coastal upwelling in the East China sea in winter, *J. Geophys. Res. Ocean.* 111 (2006) 1–11.