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Source-sink process of microplastics in watershed-estuary-offshore system

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ABSTRACT

Microplastics have been found everywhere and caused most significant pollution on earth, especially to the marine environment. However, microplastic pollution of marine environment is rarely studied based on the watershed-estuary-offshore system. In this study, a technical analysis framework of source-sink process of microplastics has been proposed based on watershed-estuary-offshore system. The impacts of human activities and hydrologic conditions on the microplastic distribution patterns in the sediments of Laizhou Bay were investigated. It is found that the river input from watershed to estuary is the main pathway for the microplastics in coastal areas to enter the sea. Population clusters, such as cities, towns and rural areas are increasingly important microplastic emission hotspots in the Laizhou Bay. Social and economic activities, productions and lifestyles in the watershed area have significant impacts on the distribution of microplastics, and the urbanization intensifies the process. A microplastic unit source-concentration response matrix model is established to simulate the transport of microplastics in the Laizhou Bay, which further confirm that river inputs are an important source of microplastics in the watershed-estuary-offshore system. It can also be concluded that the concentration of microplastics in river sediment is mainly affected by the natural conditions of the upstream watershed, the scale of human activities and the level of urbanization.

1. Introduction

Plastic pieces with the lengths smaller than 5 mm are named microplastics. Global plastic production has sharply increased with the annual output exceeding 360 million tons in 2018 (PlasticsEurope, 2019). Microplastics pollution has therefore become an urgent global environmental problem nowadays. They can directly enter the natural ecosystems from a variety of sources, such as personal care products, as primary microplastics to cause pollution (Carr et al., 2016). The microplastics from the degradation and decomposition of plastic products in nature by weathering also contributes significant pollution to the ecosystems (Browne et al., 2011; Tang et al., 2020). Up to date, microplastics have become one of the most significant pollutants on earth and microplastic pollutants have heavily accumulated in sediments around the world (Clayer et al., 2021).

Considered that microplastic pollution is a global pollution problem (Lebreton et al., 2017), initial studies of microplastics focused on freshwater (Castaneda et al., 2014; Lechner et al., 2014; Fischer et al., 2016), estuarine (Matthew et al., 2021; Carretero et al., 2021; Díaz-Jaramillo et al., 2021) and marine environments (Franciele and Campos da Rocha, 2021; Van Ryan Kristopher et al., 2021). However, the migration of microplastics from land to ocean is poorly understood (Eerkes-Medrano et al., 2015; Nel et al., 2018) Land is considered as the main source of microplastics, which would be transported to the seas by the rivers. (Lebreton et al., 2017; Schmidt et al., 2017). Rivers can carry the microplastics released by plastic manufacturing and other human activities, which is considered as a main pathway for microplastics to enter the coastal areas (Jambeck et al., 2015; Lebreton et al., 2017). In this process, rivers also become a secondary source as larger sizes of plastic break up into microplastics. (Clayer et al., 2021; Klein et al., 2015). . However, few studies have reported the microplastic abundances in rivers, showing high heterogeneity in microplastic numbers (Miller et al., 2017; Vermaire et al., 2017). Hydrological process of the watershed may be an important factor affecting the transport process of

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microplastics from land to ocean.

The particle size composition and hydrodynamic conditions of sediment can affect the settlement and resuspension of microplastics in the sedimentary environment of the semi-enclosed sea area, and ultimately change occurrence characteristics of microplastics in sediments (Rochman, 2018). Therefore, studying the marine plastic and microplastic hotspots and environmental risk assessment are significantly important (Li, 2020). Many studies have shown that human activities affect the distribution of microplastics in the intertidal zone and the bay to different extents (Peeken et al., 2018). However, the factors influencing the “source-sink” process of microplastics in the sedimentary environment of watershed-estuary-offshore system are not fully discussed.

Multivariate statistical model can be used to assess and predict the impacts of complex human activities on intertidal ecosystems. Long (2019) analyzed the historical data of sediment microplastics in the Xiamen Bay and their response to human activities, and found that the pollutants were unevenly distributed on the sea surface of the Xiamen Bay due to the influences of precipitation and tidal currents. In addition, the Shannon Index were induced to analyze the diversity and abundance of microplastics in the sediments of the coastal waters in China, and demonstrated that the distribution of microplastics from the “source” in sediment were affected by human activities (Sun et al., 2021). However, the distribution response of microplastics in watershed-estuary-offshore system to human economic activities is relatively scarce.

Watershed usually accepts the pollutants from terrestrial sources, which is greatly affected by the coastal social and economic activities (Birch et al., 2020). The rivers bring the microplastic pollutants to and drive their accumulations in the bay. The intertidal zone is the only entry of microplastics from land to sea, and thus it is the most severely affected area. Revealing the response mechanism of microplastics to human activities is very important for the development of comprehensive coastal ecosystem management strategies based on land-sea coordination. The rapid economic activities and population growth in the northern Shandong Peninsula have imposed strong pressures on the watersheds which are transferred to the coastal area by the rivers, and thus exacerbate the microplastic pollution problem in the watersheds to the Laizhou Bay. In addition, Laizhou Bay area has been undergoing rapid economic developments and population pressures for almost fifty years. The coastal area has been severely exposed to human activities,

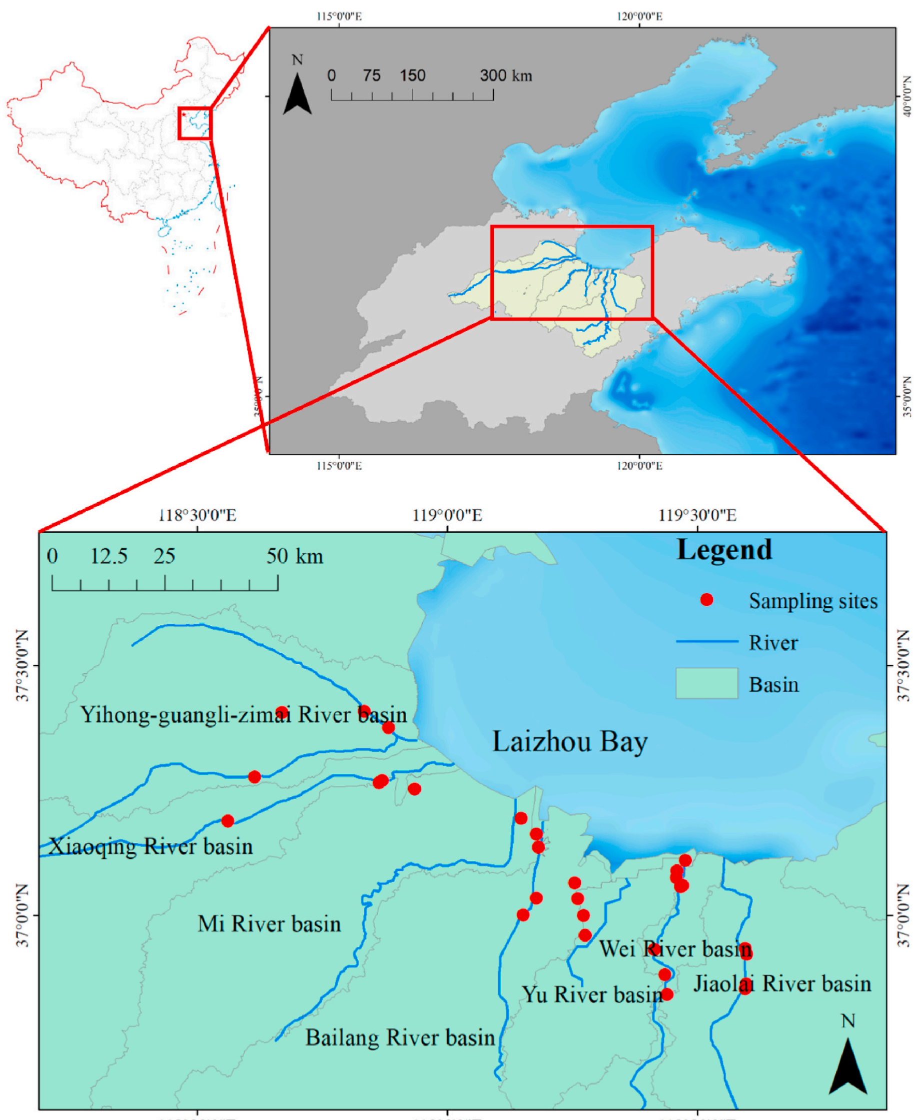

Fig. 1. Study area in Laizhou Bay. Sampling sites in eight main watersheds including Yihong River-Guangli River-Zhimai River (YHH-GLH-ZMH), Yu River (YH), Jiaolai River (JLH), Xiaoqing River (XQH), Mi River (MH), Bailang River (BLH) and Wei River (WH) of Laizhou Bay. The depths of sampling sites ranged from 0.5 m to 1 m.

especially those producing microplastics, such as waste discharge, mariculture, and local tourism. Yet the microplastics distribution patterns in the Laizhou Bay has been rarely studied based on watershed-estuary-offshore system. Therefore the knowledge gap called for the needs of this study, in which the distributions and abundances of microplastics in the sediments of eight rivers entering Laizhou Bay were studied, aiming to understand the impacts of human activities and hydrologic factors on the sediments microplastics distribution patterns in the watershed-estuary-offshore system.

2. Methods

2.1. Sediment sampling and microplastic isolation and identification

2.1.1. Sampling

Sediments in major watersheds of northern Shandong Peninsula, China were samples with a stainless-steel box sampler from 63 sites between October 30 to November 20, 2020 (Fig. 1). Based on the geographical features of the watersheds, the sampling sites were further divided into 7 groups including Yihong River-Guangli River-Zhimai River (YHH-GLH-ZMH), Yu River (YH), Jiaolai River (JLH), Xiaoqing River (XQH), Mi River (MH), Bailang River (BLH) and Wei River (WH). The depths of sampling sites ranged from 0.5 m to 1.0 m. For each sampling site, the sediment sample was collected from three randomly locations and an aluminum foil bag at −20 °C prior to analyses.

2.1.2. Microplastic extraction

The microplastics in sediment samples were extracted by the method of Thompson et al. (2004) with minor modifications. Briefly, the sediment samples were dried at 40 °C for 72 h to the constant weights. Each sample was equally divided into three portions in three glass beakers for replicate analysis. In each glass beaker, 200 g of dried sediment was mixed with 200 mL of saturated salt solution (NaCl, $\rho = 1.20$ g/mL) and manually stirred with a clean glass rod for 15 min. The sediment was allowed to settle for 5 min and carefully transferred to another glass beaker. This isolation procedure was repeated three times to increase the recovery. Subsequently, 5 mL of 30% H_2O_2 was added into each sediment suspension (200 mL) at the final concentration of 0.73% to degrade the organic matters for 12 h. The clean supernatant was filtrated through a Whatman GF/B glass microfiber filter (pore size 1.0 μm) under vacuum filtration. The glass beaker and all transfer apparatuses were rinsed several times with 200 mL Milli-Q water, and all washing solutions were filtered through the same glass-fiber filter to minimize sample loss due to adhesion of the microplastics to the walls of the filter. The filter was placed into a petri dish and air dried for microscopic inspection. Three replicates were prepared for each sampling site. During the extraction, all openings were covered with aluminum foils to prevent contamination from fibers or other particles. The extraction method was validated prior to the treatment of the collected samples for different types of polymers with different shapes. The recoveries of the microplastics are in the range of 85%–100% with the average value of 92.5%.

2.1.3. Microplastic characterization

To quantify the total number of microplastics, the filter papers were thoroughly examined under a stereoscopic microscope (Olympus, SZX10, Japan) utilizing a z-shaped pattern from left to right. Microplastics were visually identified according to the following criteria: (1) the particle cannot be broken with tweezers; (2) the particle color is evenly distributed; and (3) particles are free of tissue and cell structures (Cole et al., 2011; Hidalgo-Ruz et al., 2012). The identified microplastics were categorized into four types based the particle shapes including fibers, fragments, films and pellets. The microplastic sizes were measured on the longest sides, and then divided into the following groups: 1–1000 μm, 1001–2000 μm, 2001–3000 μm, 3001–4000 μm, and 4001–5000 μm. The smallest size of microplastic particle was found to be ~80 μm in our study. Each type of microplastic was enumerated and photographed with a microscope equipped with a camera (Cnoptec TP510, Chongqing, China).

2.1.4. Microplastic identification

A few pieces (2–5 pieces) of the microplastics found on the filter of each sampling sites with a total number of 19 microplastics were analyzed by Fourier transform infrared micro-spectroscopy (μ-FT-IR) using a Nicolet™ iN10 infrared microscope (Thermo Fisher Scientific, USA) equipped with an ultra-fast motorized stage and a single element MCT detector in the transmittance mode. The MCT detector was cooled with liquid nitrogen. Each spectrum was recorded in the range of 650–4000 cm^{-1} by co-adding 128 scans at the resolution of 8 cm^{-1}. The aperture was set to 150 × 150 μm using adjustable knife-edges. The obtained spectra were compared with the OMNIC polymer spectra library to identify the microplastics. Only the spectra with over 70% matches with the standard database were acceptable to assure the reliability of the identification.

2.1.5. Quality assurance and quality control

Strict control measures were implemented to prevent any possible contamination of the samples from the plastic products. Cotton coats, masks and polymer-free gloves should be always worn during field sampling and analysis. All instruments were rinsed three times with Milli-Q water and then dried prior to the experiments to avoid contaminations from airborne microplastics. To reduce the flow of air, the windows of lab were closed during the tests. Additionally, three procedural blanks were prepared for background correction.

2.2. Hydrologic factors and anthropogenic factors in watershed

2.2.1. Hydrological process

A hydrological analysis model is constructed with the ArcGIS modeling tools by the following main steps: filling depressions, analyzing flow directions, calculating cumulative flows, extracting river networks, generating river network nodes, and forming watershed areas.

2.2.2. Sample sites classification

The sample sites are divided into 3 levels according to their distances from the estuary of each river watershed, the distance from the river, and the elevation by the natural break point method from near to far, as shown in the Fig. 2 and Fig. S1. The watersheds are also divided into 3 levels according to the distance from the estuary, the distance of the river, and the elevation by the same method as shown in the Fig. 2.

2.3. Microplastic transport model in offshore waters

After the land-based microplastics enter the coastal areas through the estuary, they are further transported and sink in the offshore waters under the influence of hydrodynamic forces. The following section will briefly introduce the transport model of microplastics in offshore waters used for this study and its validation.

2.3.1. Model construction

The three-dimensional hydrodynamic model used in this study is constructed based on the Regional Ocean Modeling System (ROMS; Shchepetkin and McWilliams, 2005) initially configured and validated by Liu et al. (2017). It covers a wide region including the Yellow Sea and the Bohai Sea at the horizontal resolution of 1' × 1'. The model divides the vertical water column into 6 layers and includes the forces of sea surface winds, tides at the open boundary, and river flow along the coast at the hourly time step. The tidal force is derived from the Oregon State University (OSU) global inverse tidal model of TPXO7.2 (Egbert and Erofeeva, 2002), which includes four tidal constitutes, M2, S2, O1 and K1. The surface atmospheric forcing field and shortwave radiation are obtained from the Comprehensive Ocean-Atmosphere Data Set (COADS; Woodruff et al., 1998). The hydrodynamic model outputs hourly

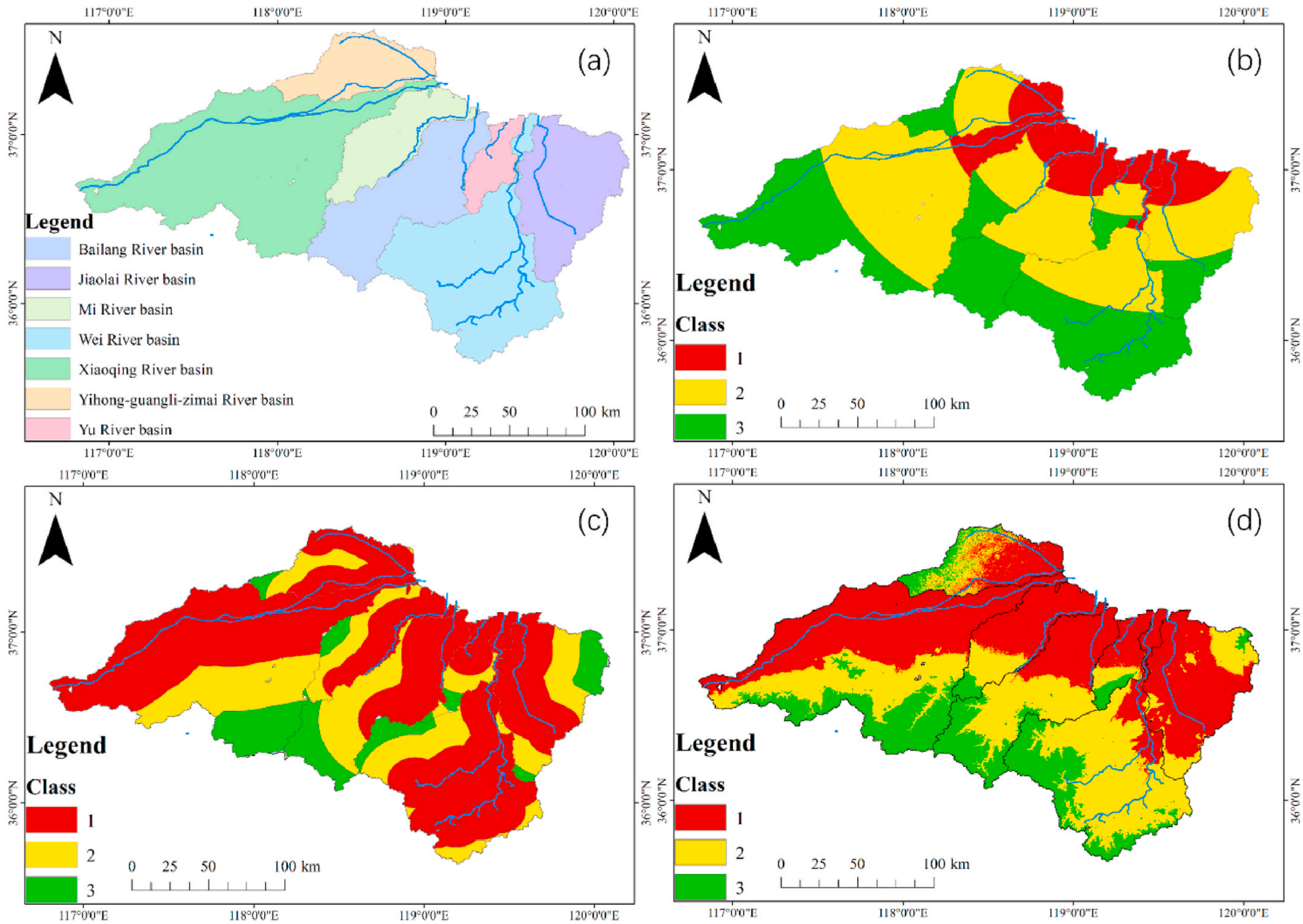

Fig. 2. Major watersheds of Laizhou Bay (a), with classifications of watersheds based on the distance from the estuary (b), the distance from the river (c) and elevation (d).

simulation results of velocities, temperature and salinity in all nodes. Based on the hydrodynamic background, a unit source-concentration response model is established to simulate the transport of the microplastics. The unit source concentration response model defines the transport of microplastics in the offshore area as the superposition of multiple pollutant point sources from the coastal rivers. The unit source concentration model is an idealized model which set the microplastic concentration of each river in proportion to the flow rate of the river along the Laizhou Bay and the transport process is simulated until the concentration becomes stable. Based on the work of Liu et al. (2017), the transport model is expressed as follows:

$$\frac{\partial C}{\partial t}+u\frac{\partial C}{\partial x}+v\frac{\partial C}{\partial y}+w\frac{\partial C}{\partial z}=\frac{\partial}{\partial x}(A_h\frac{\partial C}{\partial x})+\frac{\partial}{\partial y}(A_h\frac{\partial C}{\partial y})+\frac{\partial}{\partial z}(A_v\frac{\partial C}{\partial z})$$

where C is the concentration of microplastics; x, y and z are the components of Cartesian coordinate system representing the eastern, northern and vertical directions, respectively; u, v and w are velocities in the x, y and z directions, respectively; t is time; A_H and K_H are the horizontal and vertical turbulent diffusion coefficients, respectively; and θ_C is the point source of microplastics from the river.

2.3.2. Model validation

The unit source concentration model is a powerful tool for analyzing the transport pattern of microplastics. The unit source concentration is composed of the superposed response of several rivers as shown in Fig. 5. Land-based input via river is an important source of the microplastics in the Laizhou Bay. It is validated with the spatial pattern of microplastic concentration. Compare the Fig. 5 (a) and Fig. 6, the simulated unit source transportation presents similar pattern to the observed microplastic concentration. The high microplastic concentration cores are located around the coastal rivers. Fig. 5(b–f) are plotted in the same colormap range, and the different patterns indicate that the impact of different river input is in the different order of magnitude. Both the simulation and observation of microplastic prove that most of the microplastic in the Laizhou Bay is released through the rivers of the southwestern side of Laizhou Bay.

2.4. Multivariate statistical analysis

The attentions to microplastics in rivers have been increasing (Eerkes-Medrano et al., 2015). Recent studies indicate that there are close relationships between microplastic concentration and waste management, watershed characteristics, and hydrological conditions (Chen et al., 2020; Lv et al., 2020). MPs in the marine environment are mainly derived from agricultural pollution (Ouyang et al., 2020), riverine input (Zheng et al., 2019), sewage discharge (Akarsu et al., 2019) and maritime aquaculture (Xia et al., 2021) in terrestrial areas (Zhao et al., 2018). Therefore, this study selects the total population, population density, proportion of farmland, proportion of land for construction, GDP, consumption level, wastewater discharge, watershed area, industrial wastewater discharge, domestic wastewater discharge, general industrial solid waste volume, passenger transport volume, and nitrogen discharge as the influencing factors of the abundance of microplastics.

Some of these variables are correlated, and thus it is necessary to reduce the variable dimension, e.g. the number of variables, and identify those that can synthesize the information of the original variables. PCA is a simple, but effective method to reduce the variable dimensions, and generate new variables that can reflect the comprehensive information of the original variables, are irrelevant, and can be statistically compared. Herein, the variable dimension is reduced and new variables are generated by PCA.

The microplastic abundance is reported as the average value in the number of plastic particles per 200 g dry mass of sediment. All statistical analyses are performed using the SPSS 26.0 software (SPSS Inc., Chicago, IL, USA). The sampling locations are plotted with the ArcGIS (10.2) (ESRI, Redlands, CA, USA) software. The correlation between the abundance of microplastics and the anthropogenic factors are analyzed by the analysis of variance (ANOVA), cluster analysis and principal component analysis (PCA) with the Origin 2019 software. The positive and negative signs of Pearson's coefficient (r) represent the positive and negative correlations, respectively. The closer the absolute value is to 1, the stronger the correlation. The correlation with $p < 0.05$ is considered significant.

3. Results and discussion

3.1. Distribution pattern of microplastics in watershed sediment

3.1.1. Abundance and distribution of microplastics

The sediment analysis indicates that microplastics are widely distributed in the watersheds of Laizhou Bay, and a total number of 164 items are detected. As can be seen from Fig. S2 and Table S1, the microplastic abundances of the sampling sites in the GLH, YH, JLH, XQH, ZMH, MH, BLH, and WH watersheds vary in the range of 4–61 items/200 g of dry weight (d.w.) sediment, with the average abundance of 20.5 items/200 g d.w. The WH watershed contains the most diverse types and largest numbers of microplastics, accounting for 37% of the total number of microplastics. The mass content of microplastics in the BLH watershed is the highest, accounting for 62% of the total mass of microplastics. The microplastic amount in blue block is the highest with the values of 455 mg, which accounts for 41.4% of the total amount of all eight watersheds and 63% of the total amount of microplastics found in BLH watershed.

The occurrence, migration, and diffusion of microplastics in terrestrial and marine environments have attracted increasing attentions. Previous studies have focused on microplastic pollution in the marine environment. The latest one suggests that 70% of microplastics in the marine environment originate from rivers. The intertidal zone, river sediments, and watershed soils are important areas where microplastics sink and occur (Luo et al., 2018; Yang et al., 2020). In our study, we found large numbers of microplastics with high abundances and various sizes and components in the sediments of watersheds of Laizhou Bay. Zhao et al. (2018) also reported relatively high abundances of microplastics in the surface sediments of the Laizhou Bay and determined that the Yellow River and Xiaoqing River are the main input sources. Lin et al. (2018) demonstrated that the microplastic abundances in surface sediments from Pearl River were ranged from 80 to 9597 items/kg, which might be affected by the hydrodynamic conditions driven by multiple physical factors. It is worth noting that the sediment samples were collected from the depth ranged from 0.5 m to 1.0 m in this study, not from the surface sediment, which might cause underestimated microplastic abundances in the watershed sediment. However, the results of this study still demonstrated that microplastic pollution had widely occurred in the major watersheds of northern Shandong Peninsula.

3.1.2. Shapes and colors of microplastics

Four different types of microplastics are found in the sediments of the watersheds, including fibers, fragments, films, and pellets (Fig. 3). Films are the most common microplastics and they are present in all sediment samples collected from the eight watersheds of northern Shandong Peninsula, accounting for 35% of all microplastics. The percentages of microplastic fragments, fibers and pellets are 29%, 29% and 7%, respectively (Fig. S3). Most of the microplastics found in the eight watersheds are white (Fig. S4). Fragments are the most abundant microplastics in the JLH, BLH and MH, the microplastics found in the YH and XQH are mostly fibers. Most of the microplastics in the GLH and WH are classified as films. High abundances of microplastic pellets are only found in the ZMH.

Further analysis suggests that the microplastics are in the shapes of particle, film, fragment and fiber, among which fiber-shaped ones accounted for 93.88%. The representative microplastics were identified and analyzed. It is found that the microplastics are mainly man-made fibers and polyethylene (Zhao et al., 2018). Compared with those of other coastal areas in the world, the microplastic pollution of the surface sediments in the eight watersheds of northern Shandong Peninsula is generally at a moderate level.

The MPs carried by rivers affect the distribution of MPs in water bodies and sediment in estuarine and bay areas, and different regions

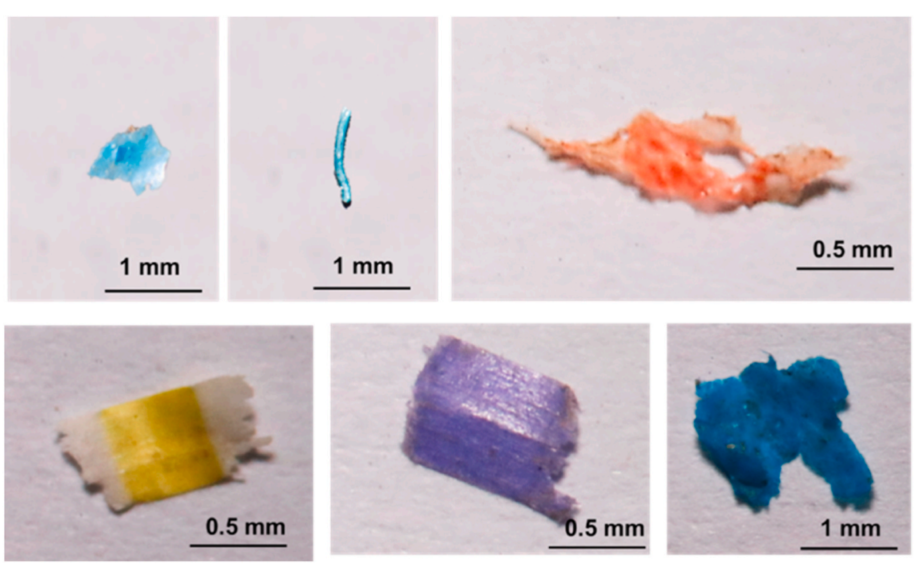

Fig. 3. Main types of microplastics in the eight watersheds of Laizhou Bay.

show different local characteristics. For example, the percentage of fibrous MPs in water bodies was 53.2% in the Qiantang River (Zhao et al., 2020), while it ranged from 17.4% to 86.7% in the Haihe River (Liu et al., 2020). The proportion of fibrous MPs in the sediment was 59.8% in Hangzhou Bay (Wang et al., 2020); however, it was 93.88% in Bohai Bay (Zhao et al., 2018). Therefore, it indicates that river input has a significant impact on the accumulation and spatial heterogeneity of MPs in the water bodies and sediment in the estuary and the bay.

3.1.3. Compositions of microplastics in Laizhou Bay

A total number of 19 particles are identified as microplastics by μ-FT-IR, which gives the identification success rate of 93.7%. The composition of other 4 particles cannot be determined because the matches of their spectra with the standard database are below 70%. Four types of polymers including polyethylene (PE), polypropylene (PP) and polyamide (PA) are found in the microplastics, and their μ-FT-IR spectra are present in Fig. S5. PE is the most abundant polymers in the microplastics. These results warn us that microplastic pollution has been widespread in the watersheds of northern Shandong Peninsula, including Laizhou Bay, and it is urgent to conduct investigations and make effective measures to prevent and reduce such pollution.

3.2. Factors influencing microplastics distributions in watersheds

3.2.1. Hydrologic analysis of watersheds

This study is based on digital elevation mode (DEM. Based on the location of the estuary and the actual situation, the study area is divided into 7 sub-watersheds from north to south and from east to west, including YLH-GLH-ZMH watershed, XQH watershed, MH watershed, BLH watershed, YRH watershed, WH watershed and JLH watershed as shown in Fig. 2. The area of the catchment area of each sub-watershed of the river flowing to the sea is calculated by equal area projection, and the results are listed in Table S2.

3.2.2. Clustering analysis

The Bray-Curtis similarity matrix is constructed after 4 extractions of the properties of 3 microplastics in 7 watersheds. The cluster analysis between the groups was conducted by the average clustering method. As shown in Fig. 2, the watersheds are clustered into three categories based on the properties of microplastics, including WH watershed, BLH watershed, and others (Fig. 2).

Twenty-three types of microplastics with the total number of 61 are found in the WH. The microplastic concentration of the BLH is the highest, which is measured to be 721.7 mg. These results indicate that the microplastic compositions and abundances of WH, BLH, and other rivers are different (Fig. 4).

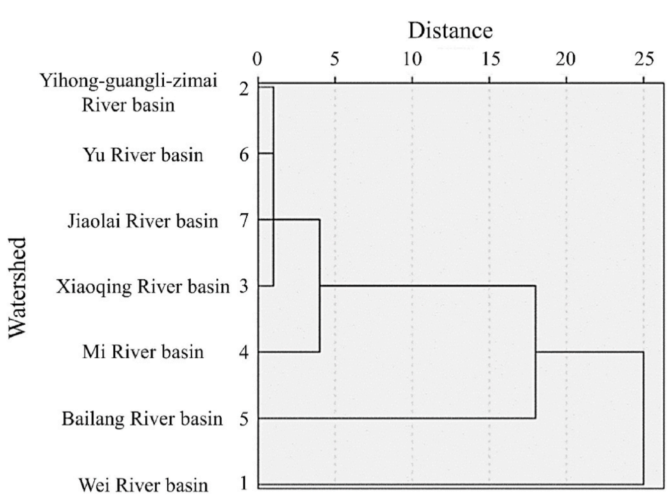

Fig. 4. Clustering analysis of watersheds based on the microplastic properties.

3.2.3. Impacts of anthropogenic factors

3.2.3.1. Principal component analysis (PCA). The impacts of anthropogenic factors are initially investigated with 16 variables. According to the classification of the watersheds, the PCA of the results is carried out. Each of the three classification methods extract three principal components. As can be seen from the variance decomposition table of the classification methods, the cumulative contribution rate of the first three eigenvalues reaches over 90%, which satisfies the criteria of selecting the number of principal components. The principle of "≥ 85%" is able to represent all the characteristics (Tables S3–8).

The principal component loadings and component matrix suggest that the extracted principal components of the three classification methods are similar. The first principal component includes population, GDP, consumption level, industrial wastewater discharge, domestic wastewater discharge, and general industrial solid waste. The principal component loadings of the main components, such as the amount of water, the area of watershed, and the amount of nitrogen release, are relatively large, indicating that the correlation coefficients between these variables and the first principal component are high and thus their impacts are significant. The first principal component is named as human activities. Rainfall and farmland ratio show relatively large loading values on the second principal component. Therefore, the second principal component is named as natural conditions. The loading of population density in the third principal component is relatively high. Population density is an important indicator of urbanization. Therefore, the third principal component is named as urbanization.

Up to date, the study on the microplastic distribution response mechanism to human economic activities is relatively insufficient. Statistical models can be used to explore and predict the impacts of complex human activities on intertidal ecosystems. Long (2019) analyzed the historical records of sediment microplastics in Xiamen Bay and their response to human activities, and found that wastes were unevenly distributed on the sea surface of the Bay and such waste distribution was affected by precipitation and tidal currents. Approximately 76.67% of the sea surface waste is generated by human activities in the watershed and surrounding land areas, and only 23.33% is generated directly by the human activities in marine, such as fishing and mariculture. The abundance of sea surface waste increases approximately as a power exponent with the decrease of the microplastic size, indicating that the miniaturization of sea surface plastic waste is obvious. The distribution characteristics of microplastics in the depositional environment can reflect the history of the plastic industry development, the evolution of the consumption structure of plastic products, and the history of important human activities in the region. Sun et al. (2021) found that the diversity of microplastics in Laizhou Bay was the highest, consistent with the diversity of pollution sources in the survey area. From the perspective of diversity, we find that human activities can influence the distribution of microplastics in the depositional environment from the "source". Meanwhile, the analysis of the influencing factors of the "sink" of microplastics in the sedimentary environment suggests that the particle size composition and hydrodynamic conditions of the sediment can affect the settlement and resuspension of microplastics, thereby mediate the redistribution of microplastics in the sedimentary environment of semi-enclosed sea areas, and ultimately change the occurrence characteristics of microplastics in the sediment.

3.2.3.2. Correlation analysis. The correlation analysis of the main components and the distribution of microplastics suggests that the number of microplastics is correlated to the second and third principal components at significant levels of $p < 0.01$ and 0.05, respectively, for the estuary distance classification method with the correlation coefficients of 0.386 and 0.307 (Table 1). The second principal component is composed of natural conditions. For the rainfall, the closer to the ocean, the greater the impact by the ocean. The stronger oceanic

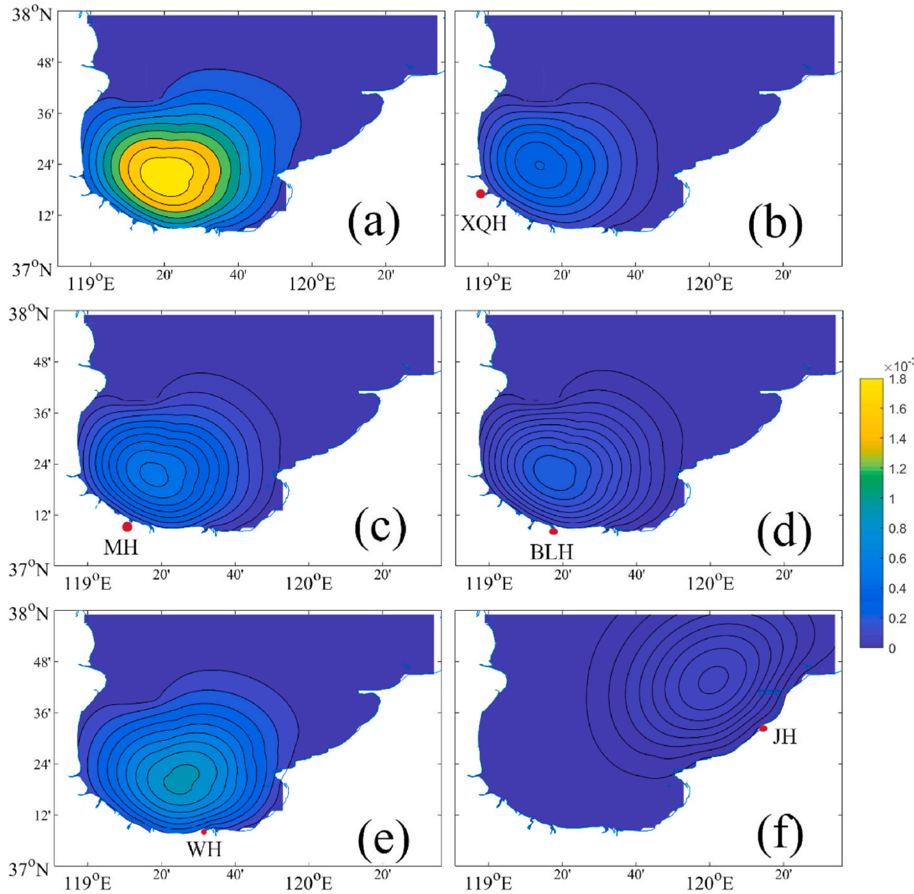

Fig. 5. Annual mean unit source concentration response of land-based microplastic loads from the coastal rivers of Laizhou Bay and the unit source response of each river. The red dot denotes the river location. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

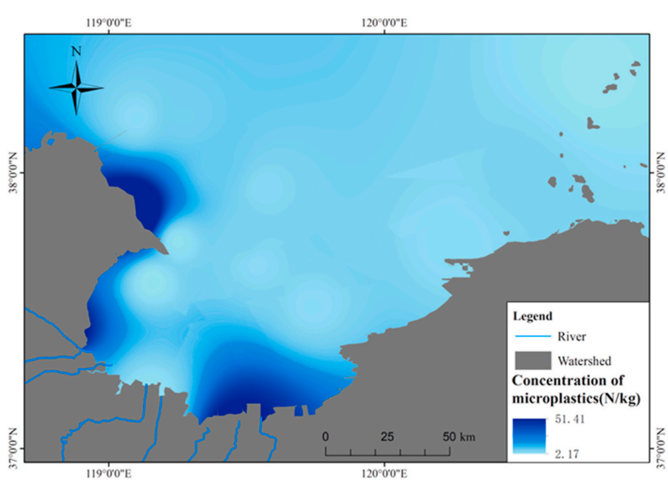

Fig. 6. Spatial pattern of microplastic concentrations from estuary to offshore.

climates and smaller daily and annual temperature changes usually result in more precipitation, which leads to more microplastics in the watershed. The third principal component represents urbanization. The areas closer the ocean have more foreign trades and higher population density, and thus are more polluted by microplastics. For the classification based on the distance from the river, the mass content of microplastics is correlated to the second principal component with the correlation coefficient of 0.436, which is significant at the level of $p < 0.05$. In the catchment area, rainwater eventually flows into the river. Therefore, the closer to the river, the more the microplastics accumulate. In the second principal component, the farmland area also shows a large loading value. Farmlands are mostly distributed near the river. Therefore, the mass content of microplastics is significantly correlated to the second principal component. For the classification based on elevation, the number of microplastics is correlated to the first principal component at the significant level of $p < 0.01$ with correlation coefficient of 0.836. The mass content of microplastics is correlated to the third principal component at the significant level of $p < 0.01$ with correlation coefficient of 0.884. Both of the first and third principal components are composed of anthropogenic factors. Plain areas with lower elevations are generally suitable for human habitation. Human activities increase the converge of microplastics.

Understanding of the micro-process mechanism of the microplastics distribution in intertidal zone and the offshore area, identifying the key influencing factors and analyzing their interaction relationship can provide theoretical supports for the sustainable development of the intertidal zone. Rishworth et al. (2016) used a generalized linear model to explore the relationship between marine stromatolite benthic microalgae community succession and the physical and chemical parameters of sea water. Later, Givan et al. (2018) assessed the impacts of human activities, species invasion, and climate change on the abundance of fish in the Eastern Mediterranean using a machine learning-based random forest model. Holon et al. (2018) was able to predict the degradation rate of Posidonia oceanica seagrass meadow ecosystem in response to different types of human activities with a random forest model, and the interpretation of model variables reached 71.3%. At present, the study on the distribution characteristics of microplastics in the intertidal zone of Laizhou Bay in response to human

Table 1
Correlation analysis of principal components and distribution of microplastics.

		Based on distance from the entrances to the sea			Based on distance from rivers			Based on distance from rivers		
		Principal component 1	Principal component 2	Principal component 3	Principal component 1	Principal component 2	Principal component 3	Principal component 1	Principal component 2	Principal component 3
Number	Pearson correlation	0.138	0.386**	0.307*	0.128	−0.337	0.187	0.836**	0.133	−0.242
	Sig. (Double tails)	0.348	0.007	0.034	0.581	0.135	0.416	0.000	0.600	0.333
Quality	Pearson correlation	0.097	0.176	−0.128	−0.025	0.436*	0.414	0.168	0.394	0.884**
	Sig. (Double tails)	0.512	0.232	0.385	0.914	0.048	0.062	0.506	0.106	0.000

Note: * represents $p < 0.05$; ** represents $p < 0.01$.

activities still lacks. Establishing of a statistical model of the Laizhou Bay ecosystem will help reveal the impacts of human activities on the changes in natural habitats and the evolution mechanism of the intertidal ecosystem.

All these indexes are consistent with the factor that the urban sites are closer to the microplastic pollution source(s) than the natural sites. Indeed, higher diversities and abundances of microplastics are expected at the areas closer to the source. In addition, the higher evenness at more distal natural sites also supports this interpretation since these sites receive the microplastic inputs and redistributes them during storms. These random-like transport processes dispatch microplastics across space and homogenize their spatial distribution.

3.3. Factors affecting offshore microplastic distribution and transport

Multiple rivers flow into the Laizhou Bay. The microplastics in Laizhou Bay are mainly brought in by the major rivers along the coast and their superimposition on each other. Fig. 5 shows the microplastic concentrations of the samples expressed as the unit source response for comparison purpose. The unit source concentration response model displays the spatial distribution of microplastics. It is implemented to assess the transport of microplastics in the Laizhou Bay. The model simulation is conducted for the data of 3 years until the results are convergent and stable. The annual mean value of the 3rd year is plotted as shown in Fig. 5. This model provides an important basis for evaluating the transportation process of the microplastics.

The unit source concentration is composed of the superposed response of several rivers as shown in Fig. 5. Land-based input via river is the key source of the microplastics in the Laizhou Bay. The microplastics will continuously influence the marine ecosystem for long time. Under the influence of the hydrodynamic processes, the high concentration core remains near the coastal region of the Laizhou Bay, indicating the land-based microplastic loads of the coastal rivers are non-negligible.

The results of unit source concentration response of land-based microplastic loads from the coastal rivers of Laizhou Bay are similar to the results of dispersion of some pollutants in Laizhou Bay, such as the reuslts of annual averaged concentration response matrixes (Dai et al., 2015), spatial distribution of water quality under the actual allocated load emissions (Su et al., 2018) and spatial distribution of inorganic nitrogen concentration in seawater under the simulation of nutrient salinity scenarios in rivers to the sea (Shi et al., 2018).

The microplastic concentrations in the estuary and offshore samples are subjected to multivariate interpolation. The microplastic concentration data of Yellow River estuary are adopted from the work of Zhou et al. (2018). The interpolation results suggest that the concentrations of microplastics near the estuaries of Yellow River, YLH, GLH, XQH, WLH, and JLH are high, with the values in the range of 51.41–23.21 items/kg, and those in the estuaries of BLH and YH are low, with the values ranging from 2.16 items/kg to 4.67 items/kg (Fig. 6). Overall, the concentrations of microplastics are high in estuaries, and gradually decreases in the offshore waters farther away from estuaries, suggesting that the rivers are the main pathway for the microplastics in Laizhou Bay to enter to the sea. Fig. 5 (a) and Fig. 6 are slightly different, but the trends are roughly the same, due to the limited number of sampling sites and errors in the simulation results.

4. Conclusions

In this study, the factors affecting the microplastic distribution patterns in the sediments of watershed-estuary-offshore system are clarified, and a microplastics unit source-concentration response matrix model is established to simulate the transportation process of microplastics in Laizhou Bay. The sediment analysis indicates that microplastics are widely distributed in the watersheds of Laizhou Bay. Fragments are the most abundant microplastics in the JLH, BLH and MH, the microplastics found in the YH and XQH are mostly fibers. Most of the microplastics in the GLH and WH are classified as films. High abundances of microplastic pellets are only found in the ZMH. In the catchment area, rainwater eventually flows into the river. Therefore, the closer to the river, the more the microplastics accumulate. Plain areas with lower elevations are generally suitable for human habitation. Human activities increase the converge of microplastics. Higher diversities and abundances of microplastics are expected at the areas closer to the source. In addition, the higher evenness at more distal natural sites also supports this interpretation. Because these sites receive the microplastic inputs and redistributes them during storms, population clusters such as cities, towns and rural areas are increasingly important microplastic emission hotspots in the Laizhou Bay. Social and economic activities, natural conditions in the upstream basin and urbanization level are the three main factors affecting the spatial distribution of micro plastics in river surface sediments. We find that human activities can influence the distribution of microplastics in the depositional environment from the "source". The unit source concentration is composed of the superposed response of several rivers, indicating that Land-based input of rivers is the main source of micro plastics in Laizhou Bay.

The superposition of the pollution loads from several rivers forms the spatial distribution pattern of microplastics. The concentration of microplastics is high in the estuary, with the values in the range of 51.41–23.21 items/kg, and gradually decreases in the offshore waters far from the estuary, which suggest that the rivers are the main pathway for the microplastics in Laizhou Bay to enter to the sea. Our work suggests that the watershed-estuary-offshore system conception model can provide an excellent analytical framework for studying the source-sink process of microplastics in coastal waters.

CRediT authorship contribution statement

Honghua Shi: Conceptualization, Methodology, Investigation, Formal analysis, Data curation, Writing – original draft, Visualization, Supervision, Project administration, Funding acquisition. **Deliang Yu:** Methodology, Validation, Writing – review & editing. **Liting Yin:** Methodology, Validation, Writing – review & editing. **Yadong Sui:** Methodology, Validation, Writing – review & editing. **Yongzhi Liu:** Methodology, Validation, Writing – review & editing. **Shuqing Qiao:** Validation, Writing – review & editing. **Weimin Wang:** Methodology, Validation, Writing – review & editing. **Wei Zheng:** Methodology, Validation, Writing – review & editing. **Dewen Ding:** Methodology, Validation, Supervision.

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

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