



Microplastics and polycyclic aromatic hydrocarbons (PAHs) in Xiamen coastal areas: Implications for anthropogenic impacts

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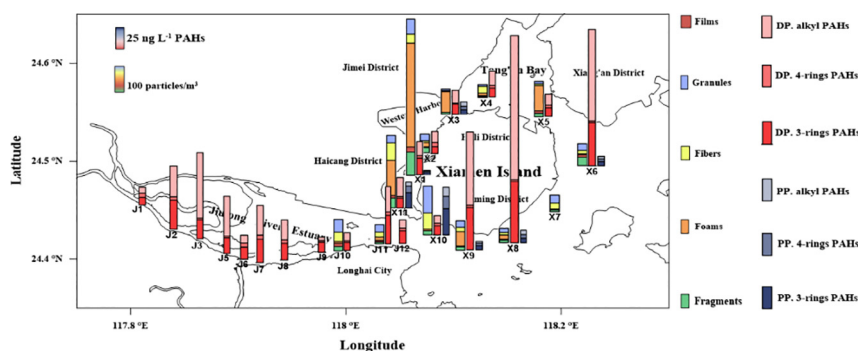
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HIGHLIGHTS

- Microplastics and PAHs were observed in the marine environments in Xiamen coastal areas.
- The abundance of microplastics showed a geographical variety, which was dominant in the Western Harbor.
- Relationships among different types of microplastics and PAHs were derived from the cluster analysis.
- Correlation analysis illustrated the possible influence of human activities on microplastics.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 29 January 2018

Received in revised form 27 March 2018

Accepted 27 March 2018

Available online 10 April 2018

Keywords:

Microplastics
POPs
Cluster analysis
Correlation analysis
Human activities
Southeast China

ABSTRACT

Microplastics and polycyclic aromatic hydrocarbons (PAHs) were investigated to study the influence of human activities and to find their possible relationship on the coastal environments, where the coastal areas around Xiamen are undergoing intensive processes of industrialization and urbanization in the southeast China. The abundance of microplastics in Xiamen coastal areas was 103 to 2017 particles/m³ in surface seawater and 76 to 333 particles/kg in sediments. Concentrations of dissolved PAHs varied from 18.1 to 248 ng/L in surface seawater. The abundances of microplastics from the Western Harbor in surface seawater and sediments were higher than those from other areas. Foams were dominated in surface seawater samples, however, no foams were found in sediments samples. The microscope selection and FTIR analysis suggested that polyethylene (PE) and polypropylene (PP) were dominant microplastics. The cluster analysis results demonstrated that fibers and granules had the similar sources, and films had considerably correlation with all types of PAHs (3 or 4-ring PAHs and alkylated PAHs). Plastic film mulch from agriculture practice might be a potential source of microplastics in study areas. Results of our study support that river runoff, watershed area, population and urbanization rate influence

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the distribution of microplastics in estuarine surface water, and the prevalence of microplastic pollution calls for monitoring microplastics at a national scale.

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1. Introduction

Plastics have been widely produced and used with enormous demands, and million tons of them finally enter into the oceans (Eubeler et al., 2010; Tosin et al., 2012). Microplastics, normally defined as plastic debris with a diameter <5 mm, originate from intentional manufacture (primary origins) or the subsequent fragmentation (secondary origins) (Fendall and Sewell, 2009; Wright et al., 2013). A large number of microplastics have been found in the seawater (Cole et al., 2011; Zobkov and Esiukova, 2017), beaches (Claessens et al., 2011; Liebezeit and Dubaish, 2012), and sediments (van Franeker et al., 2011; Van Cauwenberghe et al., 2013; Obbard et al., 2014). It has been proposed that the accumulation of floating plastic debris in different time intervals results from the combined effect of sedimentation, shore deposition, fragmentation and ingestion processes (Peng et al., 2017).

The widespread microplastics in the environment have become a hot spot since they may well exert a physical damage on the marine biota, such as internal abrasions and blockages. Moreover they have the special affinity for organic pollutants (Andrady, 2011; Wright et al., 2013), among which polycyclic aromatic hydrocarbons (PAHs) are one of the most widespread contaminants which have mainly pyrogenic sources. PAHs could pose serious risk to human health due to their toxicity, carcinogenicity and mutagenicity (Cai et al., 2016a). Therefore, microplastics play a role in aquatic ecotoxicology as vectors for these toxic substances, though the bioavailability of these pollutants carried by microplastics has not been studied in detail (Moore, 2008; Hartmann et al., 2017). The coastal or estuarine environments with mass industries and agricultural activities are considered as key areas for contaminants, such as for microplastics and organic pollutants (Browne et al., 2011; Wright et al., 2013). Compared to the beaches, seawater and sediments were the major living environment for the marine biota. Along the coastline in China, the first quantitative study of microplastics was conducted in 2013, and it focused on the seawater near the Yangtze River Estuary (Zhao et al., 2014). Besides, several studies have been reported in recent years focusing on the seawater, sediments in Yangtze River Estuary, Pearl River Estuary, Minjiang River, where massive abundance of microplastics were found (Qiu et al., 2015; Zhao et al., 2015; Peng et al., 2017).

Xiamen is a developing coastal city located in the front of the Jiulong River Estuary, the second largest river in Fujian province, China. As one of the four special economic zones, Xiamen has accomplished remarkable social and economic achievements since the early 1980s. Rapid population growth and intense industrial activities in Xiamen have led to huge pressure on coastal ecosystem stability, which attracts growing attention from the government, the public and academy (Ren et al., 2011; Ma et al., 2016; Huang et al., 2017). In addition, the Xiamen coast receives large amounts of waste from the Jiulong River Basin, where a developed agricultural region is located (Cao et al., 2005; Peng et al., 2013). This further deteriorated local ecological situation. On that account, Xiamen was select as a demonstration plot in East Asia for integrated coastal management since 1994, to explore the way to balance multiple use conflicts from both land and sea, and to control pollution sources (Cai et al., 2016b). The coastal environment situation of Xiamen coast has been the subject of extensive study and government efforts for rehabilitation. Some pollutants and their related ecological influence in Xiamen coast have been reported, such as nutrients (Cai et al., 2016b), heavy metals (Chen et al., 2010), PAHs (Ya et al., 2014), pesticide (Wu et al., 2017) and antibiotics (Zheng et al., 2011). However, there is few information about the microplastics pollution. Therefore, for effective management decision, it is urgent to investigate

the microplastics in Xiamen coast and its relationship with human activities. The aims of this study are as follows: 1) to investigate the abundance and distribution of microplastics and PAHs in Xiamen coastal areas, including Xiamen Bay and the Jiulong River Estuary. 2) to find their possible relationship by using cluster analysis. 3) to identify the sources of microplastics and see their relationship with human activities.

2. Materials and methods

2.1. Study areas

Xiamen has an area of about 1700 km² and a population of near 4 million. It is an important harbor city located on the southeast coast of Fujian Province, China. The study areas can be divided into 3 different parts (Fig. 1), i.e. the Western Harbor (including X1–5 and X11–12), the Open Sea that located at the eastern part of Xiamen Island (including X6–8), and the Jiulong River Estuary (including J1–J12 and X9–10). The Western Harbor is a dumbbell-shaped semi-enclosed bay located at the Western side of Xiamen Island (Cai et al., 2016b). The Jiulong River is the second largest river in Fujian Province, which has a basin area up to 14,700 km² and supports over 5 million populations. The open sea is located at the eastern side of Xiamen Island, where is significantly affected by the seawater outside.

Since the policy of reform and opening has been implemented for 40 years, Xiamen has become one of the most developed and populous areas in China. Intense economic activities and rapid population growth, however, have led to strong anthropogenic stress on coastal ecosystems. This area receives not only most of domestic sewage (partially untreated) and industrial wastewater from Xiamen coast, but also the agricultural runoff from the Jiulong River Basin that carries great amounts of fertilizers and pesticides.

2.2. Sampling and pretreatment for microplastics

2.2.1. Microplastics in surface seawater

All the samples were collected in March 2017 in the Western Harbor and the Jiulong River Estuary (Fig. 1). Surface seawater samples (X1–11 and J10–11) were collected using a manta trawl (1 m wide × 0.5 m vertical opening, 3 m long, 0.33 mm mesh) (Ryan et al., 2009). The net was towed a straight line with the vessel speed being kept at an average of 2 knots for 10 min (Table S2) and the top 35–50 cm of the water column was collected. A calibrated flow meter (HYDRO-BIOS, Germany) was attached to the mouth of the net to allow for volume calculation of the water filtered. The contents of the net were then washed into a pre-cleaned glass jar with Milli-Q water, and fixed in 2.5% formalin for further laboratory processes (Lattin et al., 2004).

In the laboratory, the large debris (>5 mm) were screened with a 5 mm steel-wire sieve and discarded. The rest were filtered onto nylon filters (Millipore, 20 µm), and were oxidatively conducted using 30% H₂O₂ (Nuelle et al., 2014). After filtered with nylon filters (20 µm), plastic particles were separated via flotation in NaCl solution (with a density of 1.2 g/cm³) for 24 h, and then the supernatant was collected for further analysis (Hidalgo-Ruz et al., 2012).

2.2.2. Microplastics in surface sediments

The samples were collected with a box grab at 8 stations (J1–5, J12, X3 and X12) in April 2017 (Fig. 1). And then all surface sediments samples (<5 cm) were shoveled with a clean stainless steel spoon and stored

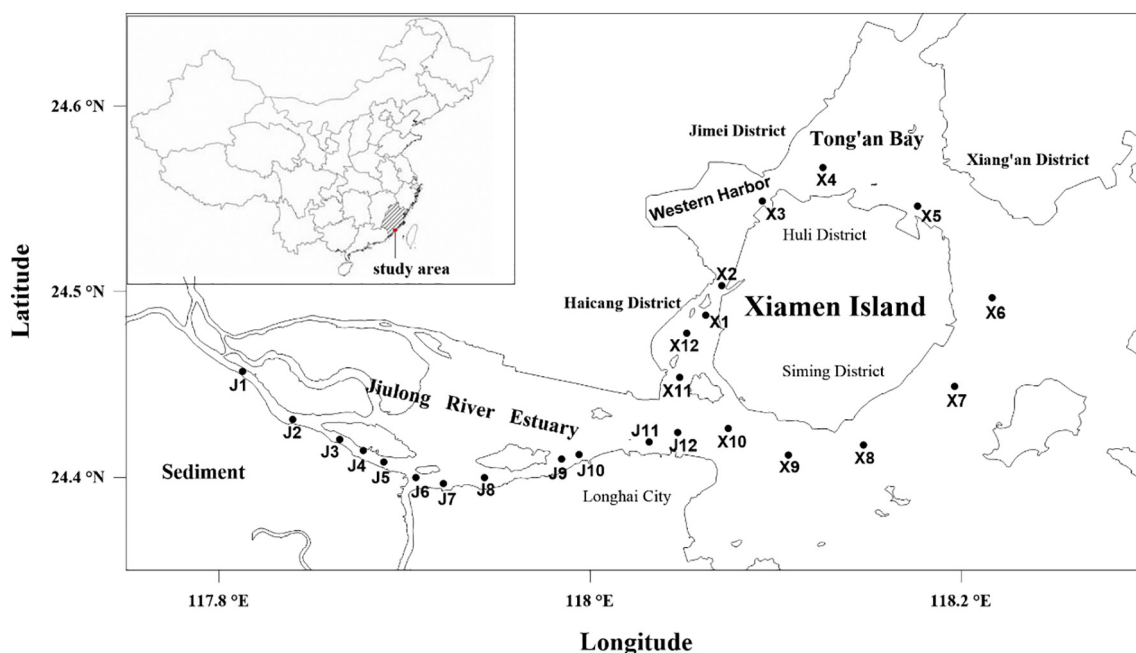


Fig. 1. Map of the sampling stations during research cruise in March and April 2017 (the details of stations are listed in Table S1). Surface seawater were collected for microplastics analysis at station X1–11 and J10–11, were collected for PAHs analysis at all stations except J4 and X7, and sediments were collected for microplastics at station J1–5, J12, X12 and X3. The salinity and temperature are shown in the Table S3.

in pre-cleaned aluminum specimen boxes until analysis. The boxes were rinsed with Milli-Q water three times at laboratory before using.

In the laboratory, the samples (500 g) were moved into 2 L pre-cleaned glass beakers and 500 mL sodium hexametaphosphate solution (51 g/L) were added to disperse the sediments by using an ultrasonic bath (30 min). The treated samples were separated by NaCl solution (with a density of 1.2 g/cm³) with a stainless steel mesh sieve (330 µm), which were assembled on the modified air flow flotation unit (Claessens et al., 2013; Zhu, 2015). 30% H₂O₂ was added to degrade organic matter for about 24 h (Nuelle et al., 2014). Then, 500 mL ZnCl₂ (with a density of 1.5 g/cm³) solution was added to glass beaker for floatation (~24 h) (Liebezeit and Dubaish, 2012). For filtration, it was poured through a nylon filter (20 µm) aided by a vacuum pump and then rinsed with Milli-Q water. Finally, the filter paper was stored in a filter paper box before microscopic observation.

2.2.3. Identification and analysis for microplastics

The filter membrane was put under a Microscope (ZEISS, Scope A1, Germany) at up to 40× magnification (Fig. 2) and taken photos using “zigzag” pattern until everywhere had been covered. If the microplastics debris were stuck together, we separated each piece with tweezers.

Microplastics were enumerated and measured by categorizing them on the basis of 5 shapes: fragments, films, foams, fibers and granules. In fact, it should be noted that foams are not a kind of shape. In our study, foams were individually picked out for counting due to their physico-chemical property. However, we listed them with the other four shapes together for clear statistics. Color was another visual characteristic we also noted, which was divided into four groups: white, black, transparent and colored. The unit of microplastics abundance was particles per cubic meter (particles/m³) for seawater, and particles per kilogram (particles/kg) for sediments. The particles were alternatively selected to identify the polymers type by using Micro Fourier Transform Infrared Spectrometer (FT-IR) (Thermo Fisher, Nicolet 6700, U.S.A). Highest similarity (at least 70% similarity for confirmation) in the database were assigned to the microplastics samples (Hidalgo-Ruz et al., 2012).

The detector spectral range was 7600–450 cm^{−1}, 16 scans at a resolution of 4 cm^{−1} (Detector DTGS). The spectra that we got were

processed by MONIC™ Picta™ software and compared with the OMNIC polymer spectra library.

2.3. Sampling and pretreatment for PAHs

All the surface seawater samples for PAHs were collected at 22 stations (J1–3, J5–12, X1–6 and X8–12) in March and April 2017. 25 L surface seawater samples in the Western Harbor and the Open Sea were collected and filtered over a GF/F filter (Waterman, USA) (0.70 µm

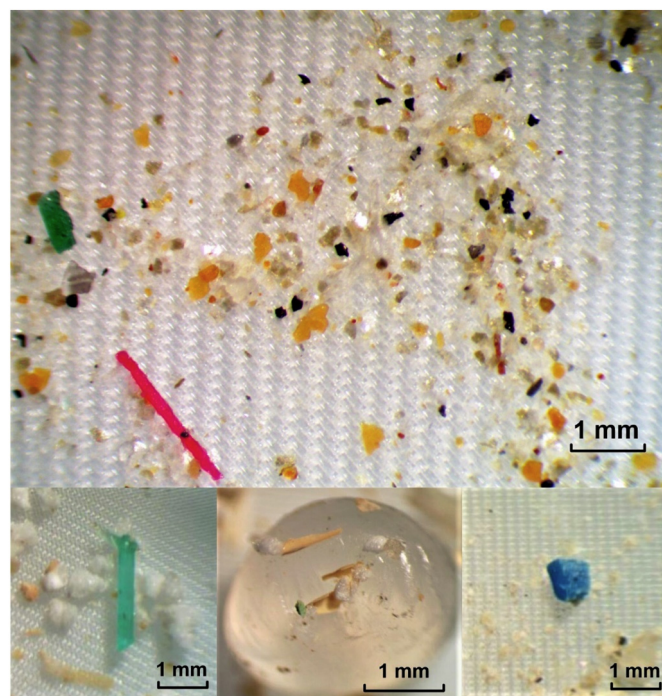


Fig. 2. The selected microplastics in surface seawater from Xiamen coastal areas under microscope (40× magnification).

pore size, 142 mm diameter) using a filtering system assisted by a peristaltic pump. The filters were collected to analyze the particulate PAHs, and 4 L of the filtered water for the dissolved PAHs analysis. In the Jiulong River, only 4 L seawater was filtered over a GF/F filter (Whatman, USA) (0.70 μm pore size, 47 mm diameter) used for the dissolved phase. The filtrate water sample was spiked with surrogate standards (acenaphthylene-d10, phenanthrene-d10, chrysene-d12 and pyrene-d12) (AccuStandard, USA), and passed through C₁₈ SPE cartridges (Envi 18, Supelco, USA) (pre-conditioned with 5 mL of methanol and 5 mL deionized water) at a flow rate of 6 mL min⁻¹.

For dissolved PAHs, the pre-treatment procedure was described in our previous paper in detail (Cai et al., 2016a; Ke et al., 2017). In brief, the targets on SPE cartridges were eluted with 10 mL of ethyl acetate (SIGMA-ALDRICH, HPLC, USA), with 8–9 g of anhydrous sodium sulfate (pre-combusted) (GB/T9853-2008) on its top to remove the water. For particulate PAHs, the freeze-dried GF/F filters were cut and spiked with surrogate standards, then extracted by acetone/n-hexane (1:1, v/v) (SIGMA-ALDRICH, HPLC, USA) in an ultrasonic bath for three times. After being merged, the extracts were concentrated to 1 mL, cleaned by pretreated alumina/silica gel chromatography, and eluted with 10 mL of dichloromethane/n-hexane (1:1, v/v) (SIGMA-ALDRICH, HPLC, USA). The eluents for both dissolved and particulate PAHs were solvent-exchanged to n-hexane and concentrated to 1 mL using a rotary evaporator. After further concentration under nitrogen and addition of internal standard (pyrene-d₁₀) (AccuStandard, USA), finally 200 μL of mixture was stored at -4 °C prior to instrumental analysis. The analysis of 16 USEPA priority PAHs and 4 alkyl-PAHs was using a gas chromatography equipped with mass spectrometry in tandem (GC-MS-MS) (the details of instrument are provided in supplementary materials).

2.4. Quality assurance and quality control

For microplastics, procedural blanks were conducted along with the samples. All of the experiment tools were rinsed at least three times with Milli-Q water and then dried before using. Lab coats and gloves were worn to avoid any contamination. Only fibers, no fragments, films, foams and granules, were found in the blanks. The blank values were low and stable, which had an average of one fibers per blank measurement (0 to 2 fibers per blank measurement). The fiber abundance reported in this study were corrected for the blanks.

For PAHs, two field blanks and three laboratory blanks were analyzed for PAHs (Table S4). Method detection limits were derived from average field blanks plus three times the standard deviation (Table S4). In addition, four perdeuterated PAHs were added to samples prior to extraction on the expedition, and the average recoveries of acenaphthylene-d10, phenanthrene-d10, chrysene-d12 and pyrene-d12 were 82 \pm 14, 72 \pm 12, 62 \pm 15 and 76 \pm 26%, respectively.

2.5. Statistical analysis of data

Cluster analysis was used to identify the environmental behaviors of microplastics and PAHs. Pearson correlation analysis was employed to investigate the relationship between microplastics abundance and various watershed characteristics (e.g., runoff, watershed area, land use) in different drainage basin. All analyses were performed by using SPSS version 22.0 (IBM Corporation).

To analyze the association between microplastics distribution and human activities, we collected related parameters of watershed characteristic and socio-economic development. The references of river runoff and watershed area, population, gross domestic product (GDP) and disposable income (DPI) and land use were listed in Table S7.

3. Results and discussion

3.1. Microplastics and PAHs in surface water

3.1.1. Abundance and distribution of microplastics

Microplastics were observed in seawater at all stations near the Xiamen coastal areas (Fig. 3 and Table S5), and the concentrations ranged from 103.0 to 2071 particles/m³, with an average of 514.3 \pm 520.0 particles/m³. In general, the extent of microplastics was in the lower level compared with those reported in seawater from Yangtze Estuary and Minjiang Estuary in China (Zhao et al., 2015; Zhao et al., 2014) (Fig. S2). Higher abundance of 4137 \pm 2461 particles/m³ and 1245 \pm 531.5 particles/m³ respectively were reported in the Yangtze Estuary and Minjiang Estuary. It should be noted that different approaches were used in those studies, thus the variation of results should be considered when compared with other studies. Except for the difference in methodology, other possible explanation of the lower abundance in the Xiamen coastal were human population-density (Browne et al., 2011) and rainy season (Ivar do Sul and Costa, 2013; Williams and Simmons, 1999). Firstly, Browne et al. (2011) found that there were more microplastics in densely populated areas with a significant relationship between its abundance and human population-density. Shanghai was one of the most populated cities in China, whose population was significantly higher than that in Xiamen and Fuzhou by up to 1 orders of magnitude. As a consequence, the abundance of microplastics in the Xiamen coastal and Minjiang Estuary were much lower than those in Yangtze River Estuary. Secondly, higher river flows in the rainy season from March to April might result in the decrease of microplastics in surface seawater. The weather of the day before sampling in March was rainy. Consequently, a significant amount of plastic debris retained in the estuary might have been washed out to the sea.

In different areas (the Western Harbor, the Jiulong River Estuary and the Open Sea), we found that samples from the Western Harbor had the highest value with an average of 695.8 \pm 735.0 particles/m³, whereas samples from the Eastern Sea and the Jiulong River Estuary had the an average of 252.0 \pm 58.90 particles/m³ and 438.8 \pm 190.7 particles/m³, respectively (Fig. 3). The abundance in surface seawater was highest in the Western Harbor between the Xiamen Island and the Haicang District, where human activities such as industrial production, bathing beach and resident areas were frequent. Geographical condition might be another reason for the highest abundance. The western harbor was a semi-enclosed bay, which had the special hydrodynamic conditions with weak seawater exchange. As a consequence, it was difficult for the floating particles to flow outside this area (Claessens et al., 2011). The lowest abundance was encountered in the open sea, and the low abundance was possibly on account of the seawater flow to the outside, which would dilute the microplastics that come from human activities at eastern Xiamen Island.

3.1.2. Shape and color of microplastics

Different shapes of microplastics were observed from the samples including fragments, films, foams, fibers and granules, and each sample area varied significantly with the others (Fig. 4A). The percentage of foams was highest in the Western Harbor (60.0%). However, foams were almost the lowest in the open sea (8.0%) and the Jiulong River Estuary (16.4%), whereas granules showed an opposite trend (10.7% for the Western Harbor, 43.9% and 46.6% for the Eastern Sea and the Jiulong River Estuary, respectively). Based on the high value of foams observed, it was possibly that at least a portion of the microplastics in the Western Harbor could be related to the point source emissions (Schilling and Zessner, 2011), such as industrial areas (Pruter, 1987; Gregory, 1991) and packaging (Derraik, 2002).

Different colors of microplastics were observed from the samples including white, black, transparent and colored (Fig. 4C). White particles were the most abundant (50%), which were dominated by the foams, followed by colored particles that account for 30%. Black and

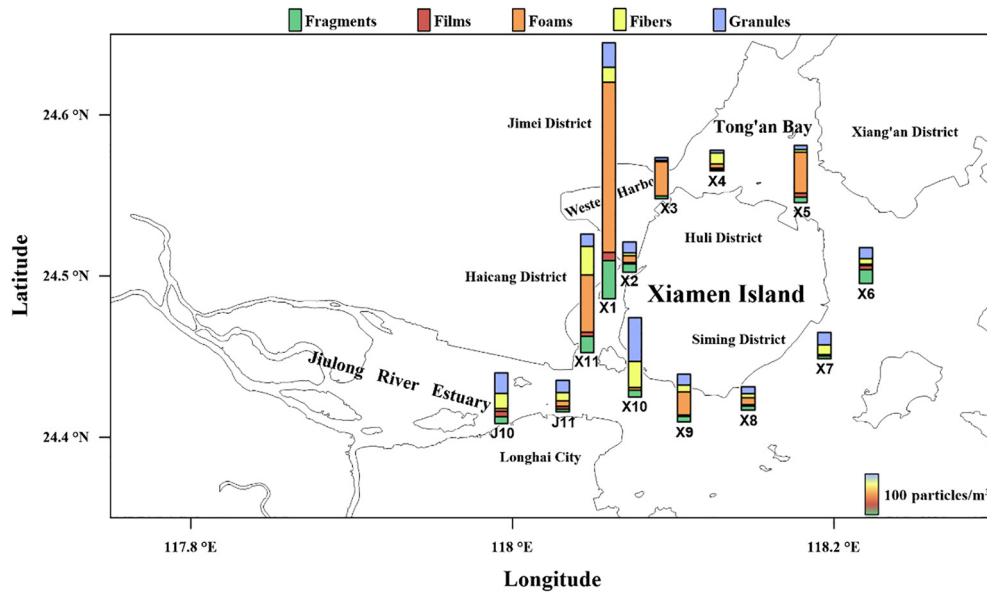


Fig. 3. Abundance of microplastics in surface seawater in Xiamen coastal area (indicated by the length of the column). Different colors represents different shapes, green represents fragments, red represents films, orange represents foams, yellow represents fibers and blue represents granules. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

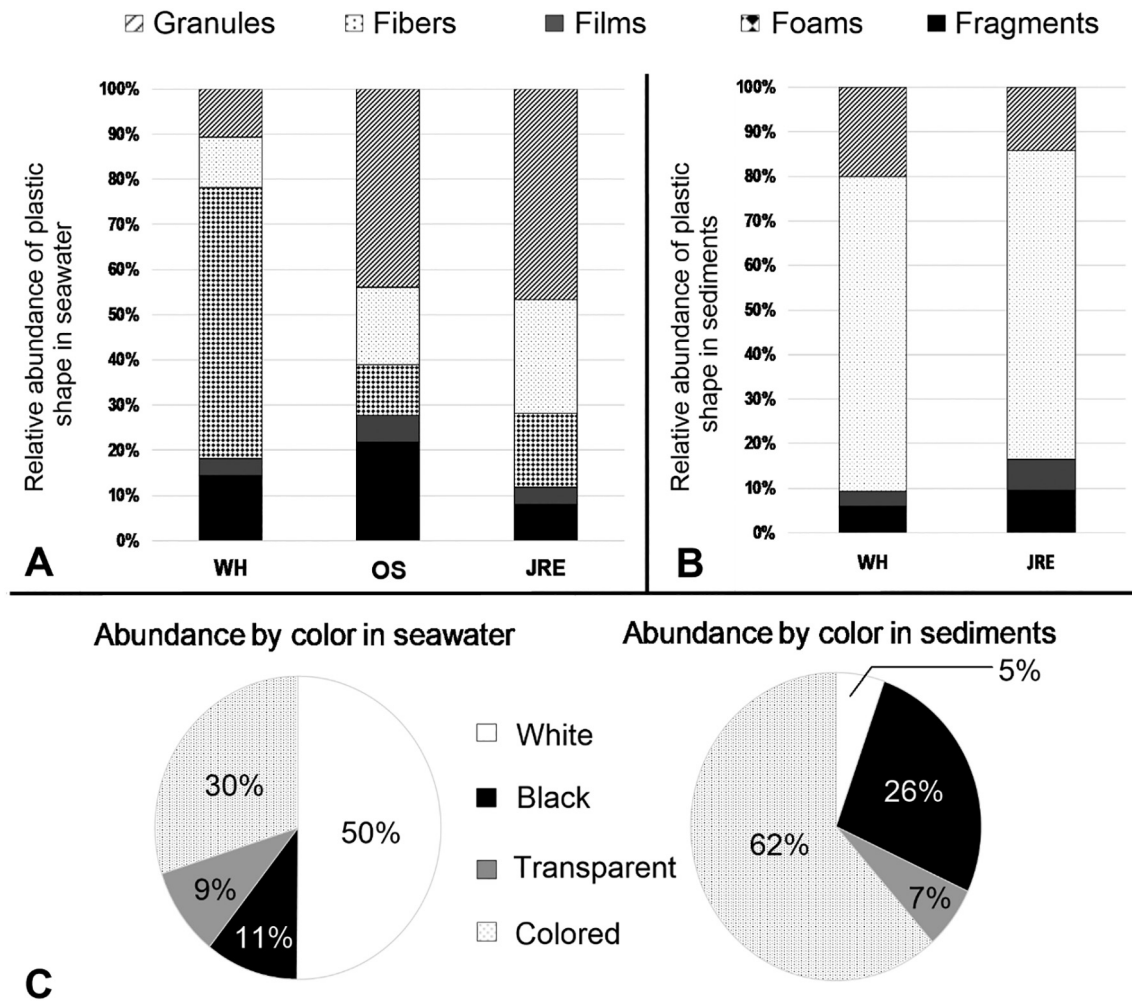


Fig. 4. (A) Composition of microplastics in seawater from different areas by shape. Results are displayed as the relative abundance of each areas. WH: the Western Harbor, OS: the open sea, JRE: the Jiulong River Estuary. (B) Composition of microplastics in sediments from different areas by shape. (C) Average relative abundance of white, black, transparent and colored particles found in seawater and sediments respectively.

transparent particles had almost the same proportion, contributing 11% and 9%, respectively (Fig. 4C).

3.1.3. Identification of microplastics

Six different polymer types were identified as microplastics in seawater samples (Table S8), including polyethylene (PE), polypropylene (PP), polystyrene (PS), polyester (PES), Polyether urethane (PU), and cellophane. All foams were identified as PS. It should be noted the presence of cellophane was not a type of microplastics. However, Peng et al. (2017) found that a part of rayon fibers in sediments of Yangtze Estuary was identified as cellophane. Rayon was a synthetic textile fiber made from cellulose (Jarmin, 1994), which differentiated them from bright color of cellophane. Because this material could not be distinguished from other microplastics during selection, and their low abundance may not affect the result significantly, cellophane was enlisted in our study.

Of all selected particles, PE and PP contributed 50.4% and 28.7% respectively, followed by cellophane (7.8%) (Table S8). The remaining proportion of particles were PES (5.2%), PU (4.3%) and PS (3.5%). There were several possible reasons that might explain the composition of materials. Firstly, the frequent use of PE and PP in industry, which were the most popular plastics in our daily life, gave rise to their large abundance (Klein et al., 2015). Secondly, the low specific densities of PE, PP and flowing PS allow for the existence in seawater rather than bottom. The low abundance of other polymers in seawater system was possibly caused by their less frequent usage and higher density.

3.1.4. Abundance and compositions of PAHs in water and SPM

The total PAHs (\sum_{13} PAHs) here included the sum of 3-ring and 4-ring parent compounds, as well as alkyl-PAHs (Table S9–12). The dissolved \sum_{13} PAHs varied from 18.13 to 247.8 ng/L (mean 61.56 ng/L), whereas the range of dissolved \sum_9 PAHs (3- and 4-ring parent PAHs) was from 12.66 to 75.32 ng/L (mean 25.86 ng/L). The 3-, 4-ring and alkyl-PAHs accounted for 41.49%, 8.57% and 49.94% of the total PAHs, respectively. It was showed that 3-ring and alkyl-PAHs were the most abundant PAHs in the surface water. The levels of PAHs in this study were higher than those in the Western Taiwan Strait (Wu et al., 2011) and Minjiang River Estuary (Zhang et al., 2004), but were comparable to those in Pearl River Estuary (Luo et al., 2008) and the Daliao River Estuary (Mohamad et al., 2011). It seemed that the concentrations of PAHs decreased gradually in recent years compared to previous researches in the same place (Maskaoui et al., 2000; Wu et al., 2017; Ya et al., 2014).

As for the spatial distribution, the abundance of \sum_{13} PAHs in the open sea was higher than that in the Western Harbor and the Jiulong River Estuary (Fig. 5), which might be attributed to frequent maritime transportation. However, high abundance detected at Station J2, J3, J5 within the Jiulong River Estuary might reflect the potential PAHs input from sewage discharges.

The abundance of \sum_{13} PAHs in particulate phase ranged from 6.16 to 57.93 ng/L (274.53–2038.9 ng/g), with a mean abundance of 19.63 ng/L (or 1044.7 ng/g). While the particulate \sum_9 PAHs ranging from 4.98 to 46.82 ng/L (or 221.39–1706.3 ng/g) with a mean of 15.38 ng/L (or 813.53 ng/g). These levels were less than those in Daliao River Estuary (Mohamad et al., 2011), but were comparable to the levels in the Pearl River Estuary (Luo et al., 2006). The highest abundance was found at station X10, which was even higher than the dissolved phase. This was just consistent with the highest abundance of SPM in this station. In terms of composition pattern of PAHs in particulate phase, the 3-ring PAHs were predominant in all SPM samples, ranging from 39.68% to 58.03% (with an average of 49.26%) of \sum_{13} PAHs. The relative content of alkyl-PAHs (23.29%) in particulate phase was lower than that in dissolved phase. In contrast, 4-ring PAHs was higher than in dissolved phase, and each percentage of them (FluA, Pyr, BaA, Chr) were similar in all selected stations, which suggested that the adsorption of 4-ring PAHs on SPM tended to be saturated.

3.1.5. Sources analysis of PAHs

In this research, PAHs isomer pairs ratios, such as Ant / (Ant + Phen), Flu / (Flu + Pyr), BaA / (BaA + Chr) and MP/P, were used as chemical tracers to reveal possible sources of PAHs (Fig. S1). Ant / (Ant + phen) ratios over 0.10 and <0.10 suggested a pyrogenic source and petrogenic source, respectively. Flu / (Flu + Pyr) ratios <0.40 indicated petrogenic source, 0.40–0.50 indicated petroleum combustion, and >0.50 indicated grass, wood, coal combustion. Besides, BaA / (BaA + Chr) <0.20 meant petrogenic sources, >0.35 meant pyrogenic sources, and 0.20–0.35 was usually taken as an indicator of mixed source (Yunker et al., 2002). What's more, MP/P ratio < 1 implied that phenanthrene was derived from pyrogenic processes, and 2–6 implied fossil-fuel sources (Prahl and Carpenter, 1983).

Fig. S1 showed the plots those isomer pairs for each samples. The results suggested that combustion sources of PAHs might contribute to the major PAHs burden in this area. The Ant / (Ant + Phen) ratios were >0.10 and the MP/P ratios were <1 for all samples, suggesting a pyrogenic source of PAHs. As for the particulate phase, the BaA / (BaA

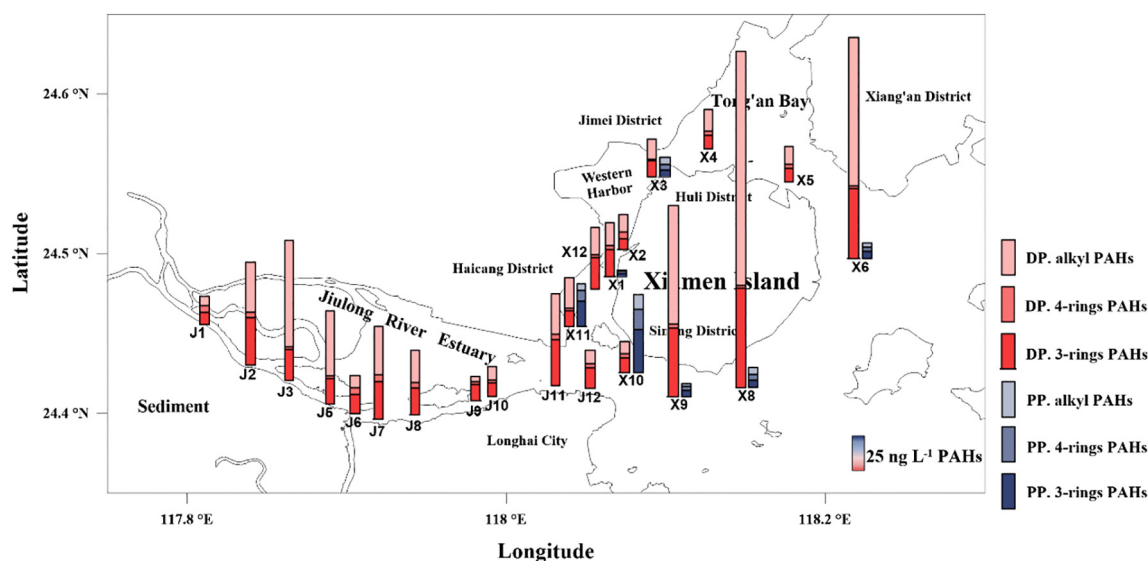


Fig. 5. Abundance of PAHs in sampling stations (indicated by the length of the column). Red and blue represents dissolved PAHs and particulate PAHs, respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

+ Chr) ratios over 0.35 and the Flu / (Flu + Pyr) ratios between 0.4 and 0.5 at all sites indicated a pyrogenic source, particularly petroleum combustion. But for the dissolved phase, the Flu / (Flu + Pyr) were >0.5 at most sites, which implied grass, wood and coal combustion source. Whereas the BaA/(BaA + Chr) ratios showed mixed petroleum and combustion-derived sources in the dissolved phase.

3.2. Microplastics in surface sediments

3.2.1. Abundance and distribution of microplastics

The number of microplastics found in sediments samples of 8 stations in the Jiulong River Estuary and the Western Harbor was presented in Fig. 5 and Table S6, respectively. The abundance of sediments samples varied from 76 to 333 particles per kilogram (particles/kg w.w.). The average microplastics abundance in all stations was 181 particles/kg. Compared with the number of sediments from Belgian coast, where the abundance was 390 particles/kg (Claessens et al., 2011) (Fig. S3), the abundance in our study areas showed a lower level. In addition, it had a much lower abundance than Kachelotplate Island and the beach in Hainan, China (Liebezeit and Dubaish, 2012; Qiu et al., 2015), which was up to 49,600 particles/kg and 6872 particles/kg to 8714 particles/kg, respectively. According to the comparison with other studies, it could be seen that the abundance of microplastics in this study was at a lower level. Notably, the differences in experimental methods should be taken into account when we compared difference of the abundance of microplastics reported in this study with other studies.

Significant difference can be found from abundance between the Western Harbor and the Jiulong River Estuary stations (Fig. 6). Sediments from the Western Harbor had the higher number with an average of 269.3 ± 80.5 particles/kg, with Jiulong River Estuary exhibiting an average of 128.0 ± 60.9 particles/m³. It was possibly because of the river input from the Jiulong River. The fact that abundance decreased along the flow direction in the Jiulong River Estuary stations can also explain the difference between two areas. Furthermore, Nor and Obbard (2014) also reported that the lowest microplastics abundance was associated with low levels of human activities. As the areas around sample stations in the Jiulong River Estuary were agricultural region, the directly discharge from human activities was negligible.

3.2.2. Shape and color of microplastics

What was different from the seawater samples is that none of the sediment samples contained foams. The other four major types of particles (based on numbers) were fibers (70.0%) and granules (17.5%), which were found at all sampling locations, as well as films and fragments account for 5.0% and 7.5%, respectively. The percentage of shapes had almost the same proportion in the Western Harbor and the Jiulong River Estuary (Fig. 4B). This indicated a possible microplastics flux from the Jiulong River Estuary to the Western Harbor. Fibrous microplastics seems to be most abundant in the marine environment, which was consistent with other findings (Wright et al., 2013). The higher proportion of fibers in sediments suggested that the microplastics were possibly derived from wastewater because of clothes washing (Browne et al., 2011). In addition, Heavy marine traffic and fishery activities was also deemed to be an important source of fibers (Browne et al., 2011).

Different from the color composition in surface seawater, colored fibers were dominant at the sediments samples. Black particles contributed to averaged 26% of all microplastics. Transparent and white particles account for 7% and 5%, respectively (Fig. 4C).

3.2.3. Identification of microplastics

Four different polymer types were identified as microplastics in sediments samples, including polypropylene (PP), polyester (PES), Poly (propylene: ethylene) and cellophane. Of all selected particles, PP was the most abundant polymer in sediments samples. However, the fact that PP was the most popular plastics in our daily life could not independently explain their existence logically on account that the density of PP was only 0.92–0.96 g/cm³. The presence of relatively buoyant microplastics in the sediments may suggest the existence of physical and/or biological processes forcing buoyant microplastics to sink to the bottom of the areas (Frere et al., 2017). And the low abundance of other polymers in sediments system was possibly caused by their high density and less frequent usage.

3.3. Classification of different types of microplastics and PAHs in surface seawater by cluster analysis (CA)

Based on the result of microplastics and PAHs in surface seawater, cluster analysis (CA) was used to further classify the different types of contaminants (fragments, films, foams, fibers, granules, 3-rings PAHs,

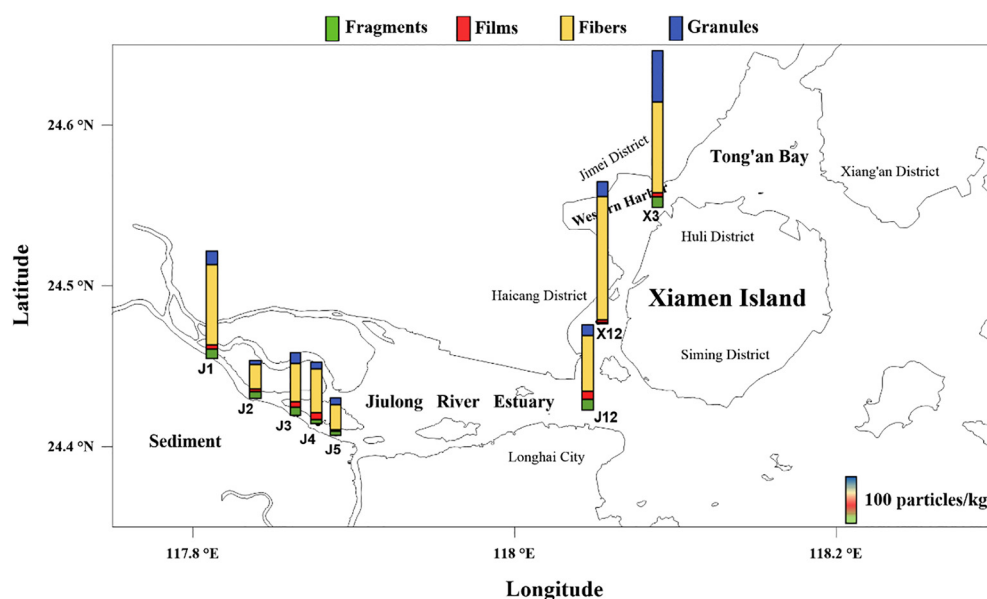


Fig. 6. Abundance of microplastics in surface sediments in Xiamen coastal area (indicated by the length of the column). Different colors represents different shapes, such as green represents fragments, red represents films, yellow represents fibers and blue represents granules. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

4-rings PAHs and alkylated PAHs) into different groups and to identify their correlations with each other. CA produced a dendrogram where the selected contaminants were divided into three clusters at Dlink/Dmax <50% (Fig. 7). The cluster 1 was only characterized with foams, which was coincident with the abovementioned analysis (3.1.2) where high abundance in the western harbor could be related to point source emissions, such as industrialized areas and packaging. Cluster 2 was characterized by fibers and granules, and it suggested that both the two types of microplastics may have the similar origins. For example, Browne et al. (2011) found that a large proportion of microplastic fibers found in the marine environment may be derived from sewage due to washing of clothes. While granules come from certain types of hand cleaners and cosmetic preparations (Claessens et al., 2011), which were also dominant at sewage. Browne et al. (2011) also found that fishing gear was also deemed as an important source of fibers. However, our result showed that fishing activities were not the major source of fibers in Xiamen coastal regions. Cluster 3 could be further divided into two parts, and films showed considerably correlation with all types of PAHs (3-rings PAHs, 4-rings PAHs, and alkylated PAHs). From our speculation, films had higher surface area than the other particles, which was conducive to adsorb PAHs. As we know, PE films were usually used to adsorb PAHs in passive sampling method (Booij et al., 2003), which was very consistent with our work. However, the specific mechanism should be the focus of future research.

3.4. Relationship between microplastics abundance and human activities

China is the largest producer and user of plastics in the world, and it may contribute the maximum proportion of waste plastic into the ocean around the world (Jambeck et al., 2015). Different levels of economic growth, urbanization, resource endowment, etc. in Chinese different areas generate apparently diversified microplastics abundance in the coastal environment. Unfortunately, the studies about microplastics pollution and available data of microplastics abundance in coastal area of China are both very limited. Here, we used the published reports by Zhao et al. (2014, 2015) with this study to discuss the possible relationship between microplastics abundance and anthropogenic activities, and to identify the potential factors for microplastics distribution in estuaries of our study areas (Table 1).

As was shown in Table 2, microplastics abundance in southeast China estuarine has positive correlation with river runoff ($r = 0.988$, p

Table 1

Characteristics of watershed proximate to estuary surface water sampled for microplastics abundance in southeast China. The references of these data were listed in Table S5.

Characteristics	Jiulong River	Ou River ^a	Min River ^a	Jiao River ^a	Yangtze River ^{b, c}
Microplastics abundance (item/m ³)	514	680	1246	956	4137
River runoff (km ³ /year)	14.06	20.20	62.37	5.17	881
Watershed area (km ²)	14,741	18,028	60,992	6603	420,237
Population (million)	9.11	6.95	16.58	4.53	234.78
Population density (km ⁻²)	618	385	272	687	559
Urbanization rate (%)	71.6	64.1	59.4	58.1	62.9
GDP per capita (RMB) ^d	78,167	54,180	54,693	46,288	67,406
DPI per capita (RMB) ^d	31,087	30,585	21,304	30,950	25,862
Farmland (%)	18.35	20.37	19.94	30.23	29.46
Forest (%)	72.85	67.44	71.82	47.45	63.06
Water area (%)	3.11	4.84	2.44	9.13	3.26
Construction land (%)	4.67	4.25	4.57	8.42	1.41
Barren land (%)	0.19	3.08	1.23	4.77	2.81

^a Microplastics abundance from (Zhao et al., 2015).

^b Microplastics abundance from (Zhao et al., 2014).

^c Since upper Yangtze River is wild and untraversed, the values of socio-economic parameters and land use only include mid-lower reach, where starts at Yibin City, Sichuan Province.

^d In view of comparability, the values of the Jiulong River in 2017 are adjusted to constant price in 2013. RMB means Chinese monetary unit.

= 0.001), watershed area ($r = 0.991$, $p = 0.001$), total population ($r = 0.986$, $p = 0.002$) and urbanization rate ($r = 0.984$, $p = 0.002$). It is not surprising that larger area and more population will lead to more waste plastics emission, and larger runoff can transport more microplastics debris from drainage to estuary. Studies in other aquatic ecosystems also support that higher urbanized or industrial areas generate more waste plastics. For example, based on the results in four estuaries of the Chesapeake Bay, Yonkos et al. (2014) observed greater concentrations of microplastics in more densely urban areas. Yangtze River estuary even has the highest concentration of microplastics in the world (Lebreton et al., 2017), because of its enormous watershed area and populations. Moreover, the defective waste management system promotes the waste plastic entering into the environment and finally reaching coastal area (Jambeck et al., 2015). It is noted that the population density has less influence on microplastics abundance.

A preliminary analysis was conducted, which was connected with the relationship between watershed economic level (GDP per capita and DPI per capita) and estuarine microplastics pollution of China, since some water quality indices have been frequently reported, such as nutrient (Chen et al., 2018) and dissolved oxygen (Wong and Lewis, 2013). The results of GDP per capita ($r = -0.415$, $p = 0.487$) and DPI per capita ($r = -0.192$, $p = 0.757$) offer a rough negative relationship without statistical significance. Part of the reason may be that the prevent awareness of microplastics pollution is not yet developed with the economic growth, so that plastic is still widely used in China (Qiu

Table 2

Pearson correlation of microplastics abundance with socio-economic and land use characteristics.

Characteristics	<i>r</i>	<i>p</i>
River runoff (km ³ /year)	0.988	0.001
Watershed area (km ²)	0.991	0.001
Population (million)	0.986	0.002
Population density (km ⁻²)	0.107	0.864
Urbanization rate (%)	0.984	0.002
GDP per capita (RMB)	-0.415	0.487
DPI per capita (RMB)	-0.192	0.757
Farmland (%)	0.604	0.280
Forest (%)	-0.114	0.855
Water area (%)	-0.258	0.675
Construction land (%)	-0.688	0.199
Barren land (%)	0.162	0.795

Pairs with *p* values below 0.01 (bold) are statistically significant.

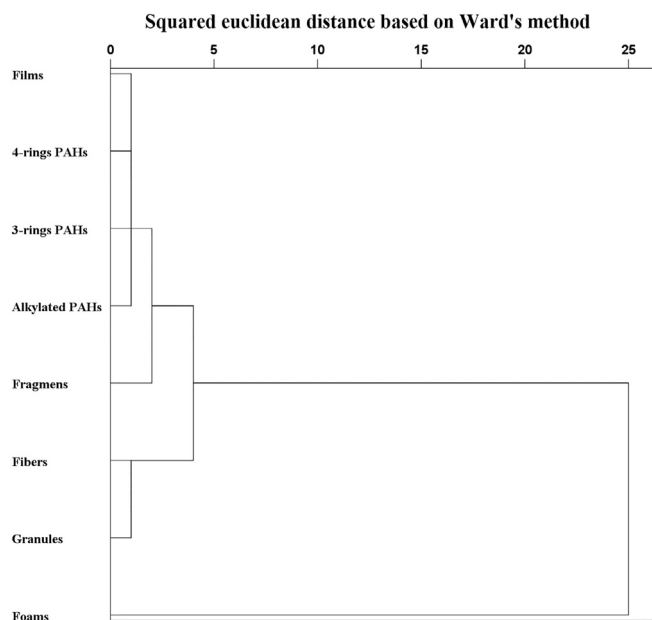


Fig. 7. Dendrogram of the microplastics and PAHs in surface seawater.

et al., 2015). In addition, there is no incentive for people to use alternatives because of the accessible, durable and low price of plastic production (Derraik, 2002).

An interesting relationship in our study was that farmland ($r = 0.604$, $p = 0.280$) was positively correlated with microplastics, while construction land ($r = -0.688$, $p = 0.199$) was on the contrary, despite lack of statistically significance. On the one hand, plastic film mulch plays an important role in Chinese agriculture due to soil warming and moisture conservation effects. Nevertheless, its residual level reaches 50–260 km/hm² in farmland based on a long-term investigation since it was hard to be recycled (Liu et al., 2014). On the other hand, more land in construction use generally means lower farmland rate, less plastic use and less residual in the environment, especially in these watersheds. Our study hints that agriculture plastic film mulch could be a significant source of plastic waste in China. On the contrary, Yonkos et al. (2014) found that more land in agriculture or forest uses generally generates less plastic waste in the US. It demonstrates that there was uncertainty in the relationship between microplastics source and land use.

4. Conclusion

Microplastics were found in all surface seawater and sediments stations. The abundance of microplastics in Xiamen coastal areas was 103 to 2017 particles/m³ in surface seawater and 76 to 333 particles/kg in sediments. The results indicated that the polluted level in surface seawater and sediments in Xiamen coastal areas was in the middle degree from all over the world. The concentrations of dissolved PAHs varied from 18.13 to 247.75 ng/L in surface seawater. Foams were dominated in surface seawater samples, however, no foams were found in sediments samples because of their low density. Microscope selection and FTIR analysis confirmed the presence of plastics in the samples and the majorities were PE and PP. The cluster analysis demonstrated that fibers and granules had the similar sources, and films had considerably correlation with all types of PAHs (3-rings PAHs, 4-rings PAHs and alkylated PAHs). Results of our study supported that river runoff, watershed area, population and urbanization rate influenced the distribution of microplastics in estuary surface water. Plastic film mulch from agriculture practice might be a potential source of microplastics in study areas.

Acknowledgement

Thanks were given to the crew of R/V Haiyang II. Financial support for this study was granted by the National Natural Science Foundation of China (NSFC) (91228202 J1210050, 40776040 and 41576180), the Natural Science Foundation of Fujian Province, China (2012J05078, 2014J06014), Senior Project of Big Science Instrument Office, Institute of Oceanology, Chinese Academy of Sciences (NO-KEXUE2016G01, KEXUE2017G09), and Xiamen University Training Program of Innovation and Entrepreneurship for Undergraduates (2016X0619, 2016X0624). We were especially thankful to Dr. Jun Wang, Ms. Jun Ye, Mr. Weimin Wang and Mr. Sadique Rayhan for their help of sampling, and Dr. Peng Huang for his assistance with data processing. We were also grateful to our reviewers for their comments and language improvements on the research.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2018.03.336>.

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