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# Characterization of microplastics in the surface seawater of the South Yellow Sea as affected by season

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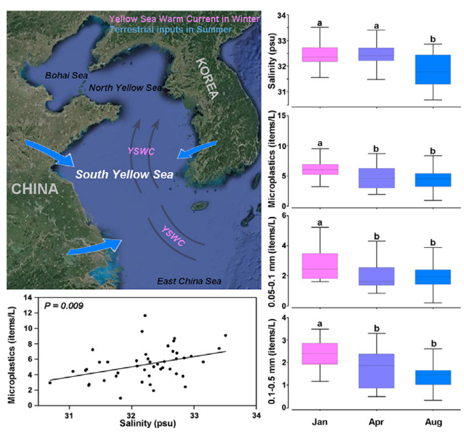
## HIGHLIGHTS

- Microplastics in the South Yellow Sea in January, April and August were investigated.
- January (winter season) had the highest abundance of microplastics (6.5 items/L).
- Microplastic characteristics in August (rainy season) had the highest variability.
- The abundances of microplastics were positively correlated with seawater salinity.
- Seasonal surveys are recommended for assessment of marine microplastic pollution.

## GRAPHICAL ABSTRACT

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## ABSTRACT

Microplastic pollution in global marine environments has attracted significant concerns; however, the environmental factors that influence the distribution and characteristics of microplastics are still unclear. In this study, 100 L of surface seawater samples collected from 16 different stations of the South Yellow Sea in January, April, and August 2018 were analyzed to investigate the relationship between spatio-temporal distribution of microplastics and environment variables. Results showed that the abundance of microplastics in January (6.5 ± 2.1 items/L) was higher than that in April (4.9 ± 2.1 items/L) and August (4.5 ± 1.8 items/L). On the whole, 78% of the total microplastics were <500 μm, ~90% were fibers, 73.2%–81.7% were transparent, and the two most abundant polymer types were polyethylene and polypropylene. The highest variability of microplastic characteristics (colour, size, and shape) and composition were observed in August, thereby showing the apparent features of terrestrial sources of microplastic pollution. Principal components analysis distinguished the spatio-temporal distribution of the microplastics, and significant difference in plastic sizes was found between microplastics in January and those in the other two months, which could be attributed to the Yellow Sea Warm Current and/or winter monsoon. Additionally, the abundance of microplastics, especially small-sized microplastics (<500 μm), was positively correlated with seawater salinity. These results suggest that microplastic pollution in surface waters of the South Yellow Sea varies with seasons owing to differences in the terrestrial sources and marine hydrological dynamics.



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## 1. Introduction

The widespread use of plastics in modern society has aroused unprecedented concern due to their ubiquity in the marine environment and their potential risks to wildlife and human health (Rochman et al., 2013). It is estimated that at least 39,000–52,000 microplastics are consumed by humans each year, and the uptake of microplastics might cause cardiovascular diseases, digestive problems, and disruptions in the reproductive system (Carbery et al., 2018; Cox et al., 2019). Over the past 70 years, there has been a constant increase in the annual global production of plastics, and the production reached 359 million tonnes in 2018 (Plastics Europe, 2019). Due to the inadequate disposal of plastics, plastic wastes continue to accumulate in the global ocean and have even been detected in polar and deep-sea organisms (Rudak et al., 2019; Taylor et al., 2016). Gradually, plastic wastes break down into plastic debris smaller than 5 mm, which are commonly referred as microplastics (Arthur et al., 2009). Owing to their small size, microplastics are readily ingested by filter-feeding zooplanktons, bivalves, and fish, and they are considered to be more dangerous to the marine ecosystem than their larger counterparts (Cole et al., 2013; Foley et al., 2018; Nadal et al., 2016). Until now, most of the studies conducted to assess microplastic pollution are based on a single-survey, and it is generally considered that the abundance of microplastics in coastal areas is higher than in remote areas that are far from human civilization (Browne et al., 2011; Wang et al., 2018). However, this view is being increasingly challenged by recent global-scale data (Barrows et al., 2018; Cózar et al., 2017; Khatmullina and Chubarenko, 2019). For example, higher or equivalent to the microplastic abundance in the sediments of the South Yellow Sea of China (560–4205 items/kg, (Wang et al., 2019a)) and the coastal seawaters of Korea (10–2000 items/m$^3$, (Song et al., 2018)) were detected in Arctic deep-sea sediments (42–6595 items/kg, (Bergmann et al., 2017)) and in the core of Arctic sea ice in the Fram Strait ($1.2 \times 10^7$ items/m$^3$, (Peeken et al., 2018)). Thus, the transportation and distribution of microplastics in marine environments is possibly influenced by other factors, in addition to human activities.

Most microplastics in oceans come from land-based sources, including riverine inputs, sewage systems, atmospheric deposition, and improper disposal (Dris et al., 2015; Klein and Fischer, 2019; Lebreton et al., 2017; Mintenig et al., 2017). Once entering the ocean, the microplastics sink to the seafloor or float on the seawater depending on their density and biological interactions. The transportation and distribution of buoyant microplastics could be influenced by water current conditions, anthropogenic activities, wind, and tides (Iwasaki et al., 2017; Rocha-Santos and Duarte, 2015). While storm surges and strong tidal waves could disturb the spatial distribution of the microplastics in sea floor sediments (Dekiff et al., 2014; Wang et al., 2019b). However, it is difficult to quantify these above-mentioned events; thus determining the relationship between microplastic pollution and environmental variables is challenging. Lima et al. (Lima et al., 2014) reported that microplastic abundance in the Goiana Estuary was positively correlated with dissolved oxygen and salinity. However, another study in the Atlantic Ocean found no correlation between microplastic abundance and environmental parameters, including chlorophyll, pH, salinity, and wind speed (La Daana et al., 2017). Nevertheless, quantitative data on oceanic conditions obtained via surveys is crucial because it provides a better understanding of the potential transportation mechanism of marine microplastics.

The South Yellow Sea is a semi-closed epicontinental sea bounded by Chinese mainland and the Korean Peninsula. Three important industrial and economic provinces, including Shandong, Zhejiang, and Jiangsu Provinces, lie along its coast, and significant amount of domestic sewage is released into the marine environment. This sea region is influenced by the East Asian Winter Monsoon, Yellow Sea Warm Current, and riverine input (Liu et al., 2015; Zhang et al., 2008). However, there is so far no report on the temporal and spatial distribution of microplastics in the South Yellow Sea by conducting multiple surveys, let alone the relationship between microplastic pollution and environmental variables. Therefore, this study aims to investigate the spatio-temporal variations in the abundance, distribution, and characteristics of microplastics in the surface waters of the South Yellow Sea based on three surveys that were conducted within a year. Moreover, the relationship between the spatio-temporal patterns and environmental variables was investigated. These results will provide a better understanding of the distribution mechanism of microplastic pollution in the South Yellow Sea.

## 2. Materials and methods

### 2.1. Sample collection

In this study, 16 sampling stations were set up in the South Yellow Sea (Fig. 1, Table S1). In January, April, and August 2018, 100 L of surface seawater (0–50 cm) was pumped continuously from each station at a flow rate of 40 L/min through a long pipe to the deck when the research vessel (Dong Fang Hong II) was stopped. Of the three sampling months, January falls within the winter dry season, and August lies within the summer rainy season. Due to limited accessibility, station H06, H02, and H12 were unfortunately omitted for January, April, and August. Seawater temperature and salinity at each station were recorded using a SBE19-CTD profiler (Sea-Bird Electronics, WA, USA, Table S2).

The seawater was successively filtered through a 5-mm stainless-steel mesh and a 50-μm plankton net. The plankton net was then rinsed with the filtered seawater, and the detained items were collected in 500 mL glass bottles, and stored at 4 °C prior to microplastic analysis.

### 2.2. Microplastic isolation

In the laboratory, 50 mL of hydrogen peroxide ($H_2O_2$, 30%) was added into each glass bottle and maintained at room temperature for 24 h to digest the biological materials (Tamminga et al., 2019). Thereafter, 300 mL of zinc chloride ($ZnCl_2$, approximately 1.6 g/mL) solution was added to suspend the microplastics (Bergmann et al., 2017). After 12 h, seawater samples were carefully poured into stainless-steel sieves with mesh sizes of 2, 1, 0.5, 0.1, and 0.05 mm, and the particles detained on each sieve were classified into different size groups, i.e., 2–5 mm, 1–2 mm, 0.5–1 mm, 0.1–0.5 mm, and 0.05–0.1 mm, respectively. To reduce microplastic loss, the above procedures were repeated thrice. Finally, the detained particles on each sieve were washed with distilled water into petri dishes (d = 10 cm), and placed in an incubator at 60 °C for 24 h.

### 2.3. Contamination protection and quality control

To minimize artificial and airborne contamination, the sieves, glass beakers, and petri dishes were rinsed with 0.45-μm filtered distilled water and covered with aluminum foil before use. Before using the prepared $ZnCl_2$ solution to suspend the microplastics, it was filtered using a 5-μm membrane filter (Millipore). During all the procedures, cotton laboratory coats and latex gloves were worn to reduce synthetic textile contamination. Additionally, to further prevent contamination, the observations were conducted by one person in a closed room. To evaluate possible contamination, three procedural blanks (0.45-μm-filtered seawater, 10 L) were filtered and analyzed simultaneously (Cincinelli et al., 2017), and only one suspected microplastic was observed in all three blank controls.

### 2.4. Identification of microplastics

The samples stored in petri dishes were visually classified, and their sizes, colors, and shapes were observed using a stereo microscope with 20–80× magnification (SZN71-B4, Sunny Optical Technology, Zhejiang, China) and identified according to pre-set criteria (Hidalgo-Ruz et al., 2012), including no cellular or organic structures, becoming sticky under hot needle test, and presenting clear and homogeneous colors. The observed fibers were defined as microplastics if they had a slender,

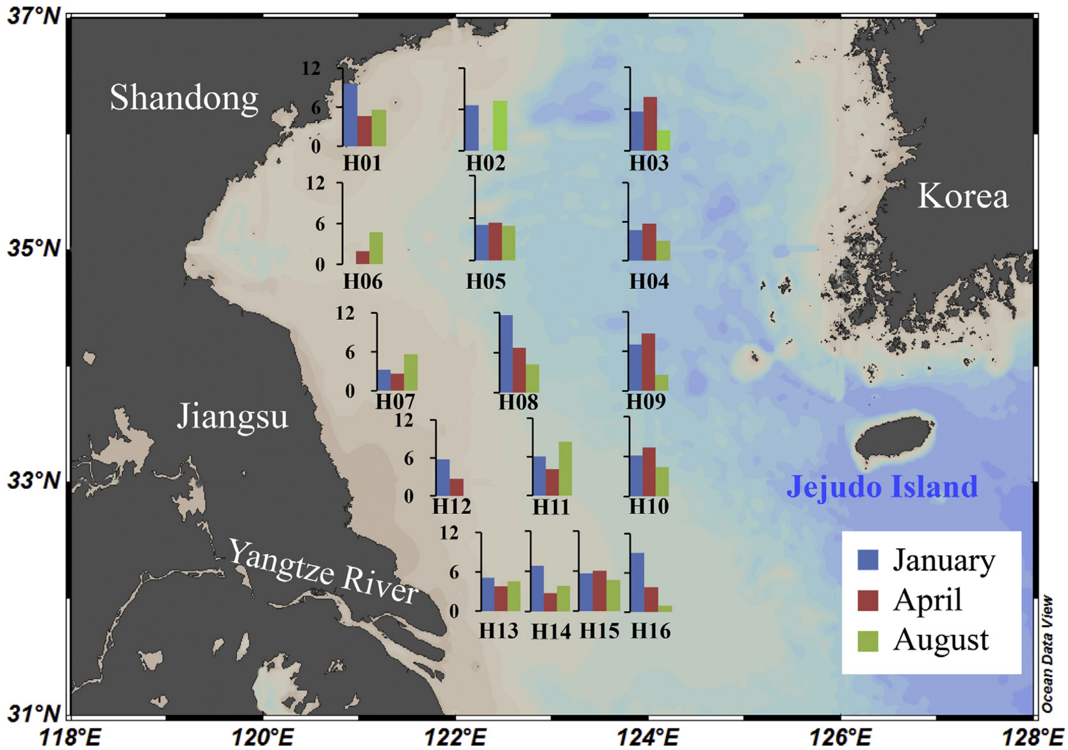


**Fig. 1.** Sampling stations and abundances of microplastics in surface seawater in the South Yellow Sea in January, April, and August 2018. The ordinate indicates the abundance of microplastics (items/L).

bent, and greatly elongated appearance, while lines had relatively thick and straight borders. After visual identification, randomly selected items (>1 mm) were further analyzed using Fourier transform-infrared spectroscopy (FT-IR) under the attenuated total reflection module. The spectra were obtained using Nicolet Nexus 470 FT-IR spectrometer (Thermo Scientific, Madison, WI, USA) at the measurement resolution of 4 $cm^{-1}$ and wave range of 650–4000 $cm^{-1}$, respectively. Then, the spectra obtained were compared with those in the software database, and a similarity >70% was accepted as a polymer match.

### 2.5. Statistical analysis

The abundance of microplastics in seawater was expressed as number of items per liter of seawater (items/L). The temporal distribution pattern of the microplastics at different sampling stations was determined via principal component analysis (PCA) using PRIMER v6.1 package (Clarke and Gorley, 2006) based on log-transformed/normalized data. Differences among groups of samples were tested using permutational multivariate analysis of variance (PERMANOVA), which is a non-parametric multivariate statistical test that is used to compare groups of objects and test the null hypothesis that the centroids and dispersion of the groups as defined by the measure space are equivalent for all groups (Anderson et al., 2008). Linear regression analyses of the spatial distribution of the microplastics in relation to salinity/temperature were performed using SigmaPlot v10.0, and $p < 0.05$ was considered statistically significant. Additionally, a geographical map was created using the software ODV (R. Schlitzer, Ocean Data View, 2003, http://www.awi-bremerhaven.de/GEO/ODV).

## 3. Results

### 3.1. Microplastic abundance

Microplastics were detected in all the seawater samples, and they could easily be classified into five main shapes (Fig. S1). The total number of microplastics in January, April, and August was 9808, 6890, and 6748, with average abundances of 6.5 ± 2.1, 4.9 ± 2.1, and 4.5 ± 1.8 items/L, respectively (Fig. 1). The maximum microplastic abundance was observed in the sample collected at station H08 (11.7 items/L, Fig. S2) in January, and station H16 in August had the minimum abundance (0.96 items/L, Fig. S3). In April, it was observed that the abundance of microplastics ranged between 1.9 and 8.7 items/L (Fig. S4). The ratios of the maximum abundance to the minimum abundance of microplastics observed at the different stations in January, April, and August were 3.6, 4.5, and 8.7, respectively.

### 3.2. Microplastics characteristics

The most frequently observed microplastics had sizes <500 μm, and they accounted for approximately 79%, 78%, and 76% of all the microplastics in January, April, and August, respectively (Fig. 2). Compared with April and August, January had the highest percentage of 1–2 mm microplastics (9.6%) and the lowest percentage of 2–5 mm microplastics (4.0%). The percentages of microplastics in the size range of 2–5 mm in April and August were 7.7% and 6.8%, respectively. The size distribution of microplastics at each station showed a similar trend for the different sampling months, especially in April and August (Figs. S5–S7). Transparent microplastics accounted for 81.7, 73.9, and 73.2% of the total identified particles in the seawater in January, April, and August, respectively. By contrast, microplastics observed in August had the highest percentages of yellow (3.4%), blue (14.4%), and red (3.7%) particles. For each station, the microplastics observed in April and August also showed a similar colour distribution (Figs. S8–S10). Fiber-shaped microplastics were most predominant, and accounted for 91.3%, 93.1%, and 89.3% of the total particles in January, April, and August, respectively (Figs. S11–S13). Compared to January, August had higher percentages line (7.1%), granule (1.2), film (0.79%), and fragment (1.6%) microplastics.

FT-IR analysis results showed that polyethylene (PE) and polypropylene (PP) were the two most common microplastic types (Fig. 3).

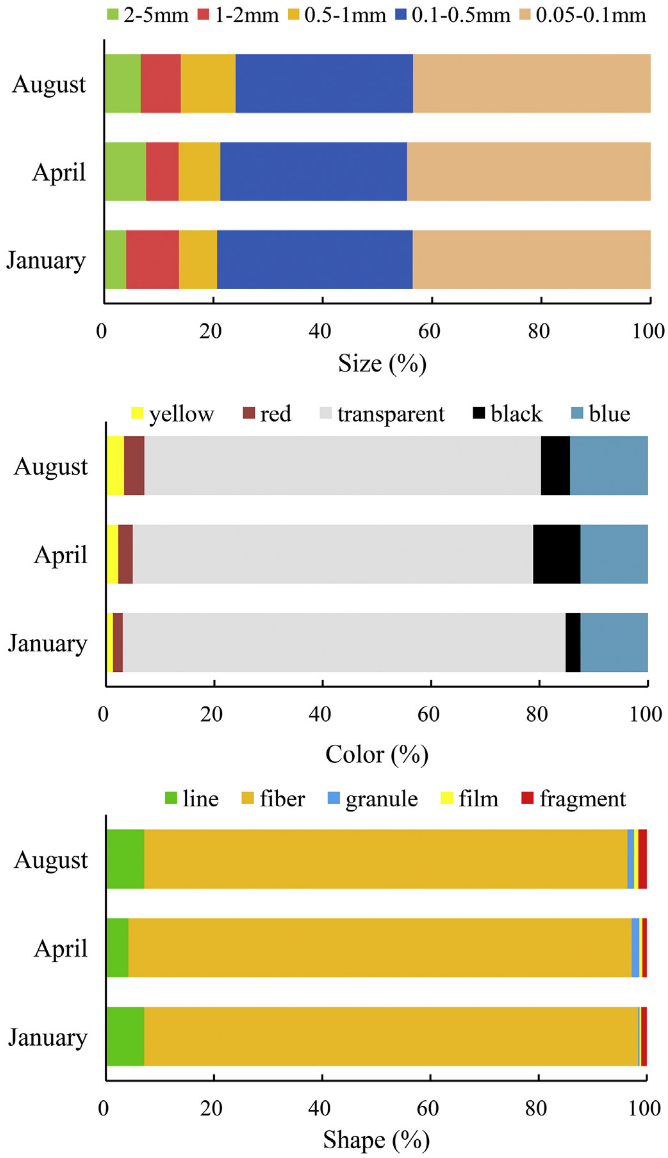


**Fig. 2.** Size, color, and shape distribution of microplastics in surface water collected from the South Yellow Sea in January, April, and August of 2018.

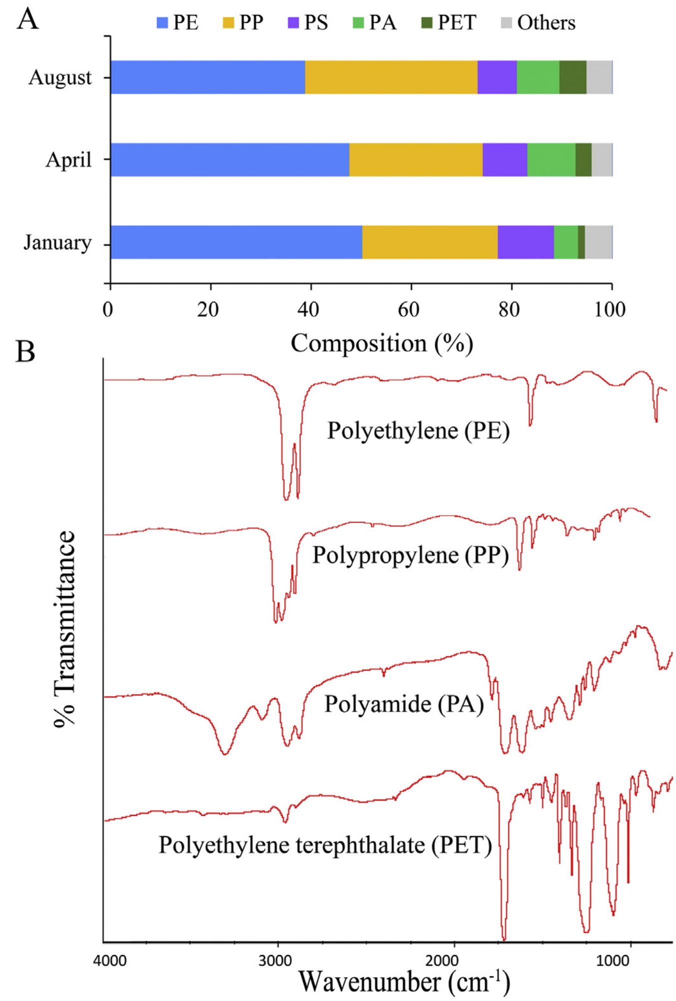


**Fig. 3.** Compositions (A) and FT-IR spectra (B) of microplastics separated from the seawater in the South Yellow Sea in January, April, and August of 2018.

PE accounted for 50.2%, 47.6%, and 38.8% of the total microplastics in January, April, and August, respectively. August had the highest composition of PP, polyamide (PA), and polyethylene terephthalate (PET) polymer types than that in January and April.

### 3.3. Spatial pattern in size spectrum of microplastics

PCA using 46 microplastic density data with size spectra from three sampling periods are shown in Fig. 4. The two PCA axes in the plot explained a significant proportion (70.6%) of the total microplastic variability and separated the January samples from those of April and August. Furthermore, the PERMANOVA test revealed clear differences among sample clouds from the three sampling periods (pseudo-$F$ = 2.129, $P = 0.047$), and significant differences between January and the other two months were also observed using pair-wise tests ($P < 0.05$, Table S3). Notably, the vectors of small-sized microplastics (0.05–0.1 mm, 0.1–0.5 mm) pointed toward the January samples, while the vectors of larger-sized microplastics pointed toward the April and August samples.

The microplastic abundances showed significantly positive correlations with the salinity ($P = 0.009$, Fig. 5A); however, no correlation

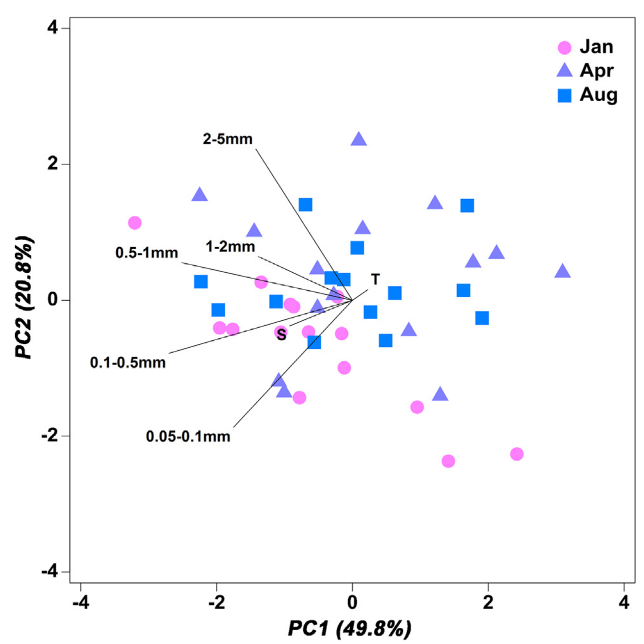


**Fig. 4.** Principal component analysis plots based on log-transformed microplastic data for temporal distribution from the South Yellow Sea in January, April, and August of 2018.

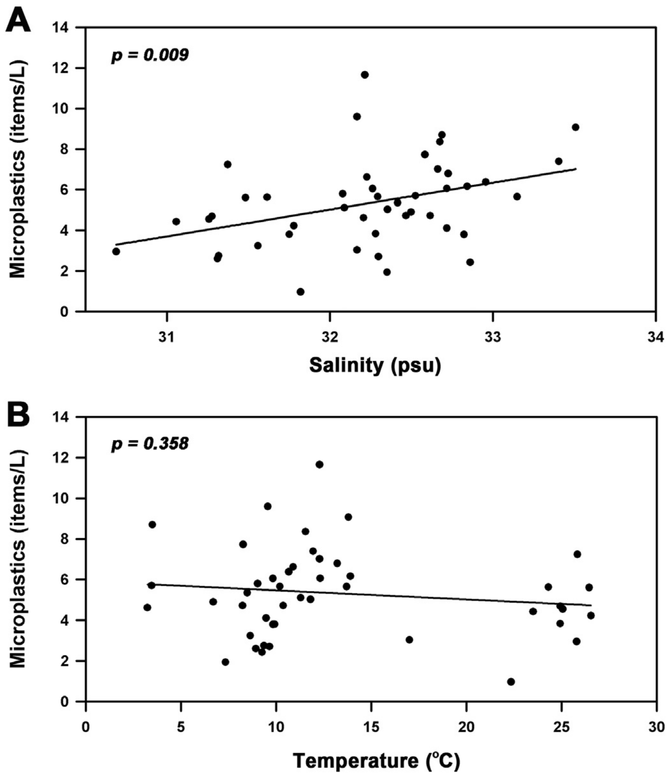


**Fig. 5.** The relationship between microplastic abundance (items/L) and salinity (A)/temperature (B) of seawater from the South Yellow Sea was analyzed by scatter plots with fitted linear regression lines.

with temperature was observed ($P = 0.358$, Fig. 5B). It is worth noting that the abundances of microplastics with sizes in the ranges of 0.05–0.1 mm and 0.1–0.5 mm also showed ahad strong correlation with salinity ($P < 0.05$, Fig. S14).

In January, the plots of salinity and microplastics abundance overlapped the most arear and showed a similar distribution pattern in their contour maps. The values of salinity and microplastic abundance in the western part of sea were low, while those in the southeast part were obviously higher (Fig. 6A, B). In April, some areas showed high levels of salinity and microplastic abundance, and these areas were located midway between Jejudo Island (South Korea) and Jiangsu Province (China, Fig. 6C, D). However, the contour maps of salinity and microplastic abundance in August did not show a similar distribution pattern (Fig. 6E, F).

## 4. Discussion

The study investigated the spatio-temporal distribution and characteristics of microplastics in the seawater of the South Yellow Sea and moreover found that microplastic pollution showed a positive correlation with seawater salinity. The Asian sea areas are regarded as microplastic hot spots and have attracted considerable attention (Isobe et al., 2015; Jang et al., 2017; Li et al., 2018). The average abundance of microplastics observed based on the three surveys was 5.3 items/L, which is similar to the results of Chinese estuary and coast waters (0.01–11 items/L, Table S5; (Zhang et al., 2017; Zhao et al., 2014)) and nearshore areas of South Korea (0.01–2 items/L, (Song et al., 2018)), but far lower than that in surface seawaters of the Incheon/Kyeonggi Coastal Region (152.7 ± 92.4 items/L, (Chae et al., 2015)). A high abundance of microplastics in seawater possibly increases the probability of microplastic ingestion by commercially important marine species, resulting in seafood safety and quality problems (Barboza et al., 2018; Hantoro et al., 2019). Among the three months, January had the highest microplastic abundance (6.5 items/L) and lowest spatial variance of the ratio of the maximum and minimum abundance of microplastics among stations (3.6) was observed. By contrast, August had the lowest microplastic abundance (4.5 items/L) and the highest spatial variance (ratio = 8.7). Surprisingly, most previous studies have solely focused on the spatial distribution of marine microplastics and neglected the temporal distribution (Lorenz et al., 2019; Rodrigues et al., 2018). A study in the Northwestern Mediterranean Sea revealed that at the same location, microplastic abundance changed by a factor of 4 within two consecutive days (Constant et al., 2018). For seasonal comparisons, Cheung et al. (Cheung et al., 2018) reported that microplastics in the Pearl River Estuary were significantly more abundant in the rainy season (July) than that in the dry season (February). In this study, microplastic abundance in the dry season (January) was 45% higher than that in the rainy season (August), which does not agree with the findings of these above studies. Similarly, Rodrigues et al. (Rodrigues et al., 2018) found that the abundance of microplastics in the water of the Antuã River (Portugal) in the dry season (October) were significantly higher than that in the rainy season. Thus, temporal distribution of microplastic pollution may be limited owing to significant changes in the microplastic abundance under different environmental situations. Additionally, atmospheric deposition has been considered as an important pathway for introducing microplastics in the ocean, and small-sized fiber- and fragment-shaped microplastics (<500 μm) have been identified as the main forms of plastic in the atmosphere (Allen et al., 2019; Liu et al., 2019). In this study, small-sized and fiber-shaped microplastics were most predominant, especially in January. Thus, we suspected that heavy air pollution during the winter dry season (January) possibly resulted in an increase in the transportation of microplastics from land to ocean. In the rainy season (August), surface runoff could transfer microplastics to the coastal areas, but many environmental parameters including hydrodynamic conditions, wind, tide, and salinity may affect the behavior and distribution of the microplastics in marine environments (La Daana et al., 2018). Therefore, it is necessary to investigate the spatio-temporal distribution of microplastics to better understand the level of microplastic pollution.

In this study, the main size range of the microplastics observed was 50–500 μm (>75%), which is slightly lower than that reported in a previous study in the same region (90%, (Sun et al., 2018)). Size is an important feature that influences the potential ecological risks of microplastics. The detection of a large number of small-sized microplastics might indicate an extreme threat to marine organisms due to the high level of ingestion and adsorption capacity of these hazardous pollutants (Auta et al., 2017). In this study, fibers were identified as the most predominant form of microplastics throughout the three sampling months, a finding that is consistent with other studies in the Yellow Sea (Sun et al., 2018; Zhao et al., 2018). January had the highest proportion of transparent microplastics (81.7%), while the microplastics observed in August showed higher proportions of microplastics with diverse colors and shapes. Additionally, the proportion of microplastics with sizes in the range of 2–5 mm observed in the months of April and August was higher relative to January. Interestingly, in the rainy season, particularly August, the lowest microplastic abundance and the highest variability in the microplastic characteristics were observed. During the rainy season, surface waters could transport microplastics from the land into the ocean. In East Asia, approximately 74% of the all the microplastic release occurred during the rainy season (May to October, (Lebreton et al., 2017)). Therefore, surface runoff might transfer microplastics into the marine environment and thus increase the microplastic diversity.

This study also revealed that fewer PE, and more PP, PA PET microplastics were detected in August compared to January. Even though the distribution of the polymer types observed in this study was similar to that observed in other studies in the Yellow Sea (Sun et al., 2018; Zhu et al., 2018), the changes in the distribution of polymer types showed apparent features of terrestrial sources. PP is a thermoplastic with both industrial and household applications, including

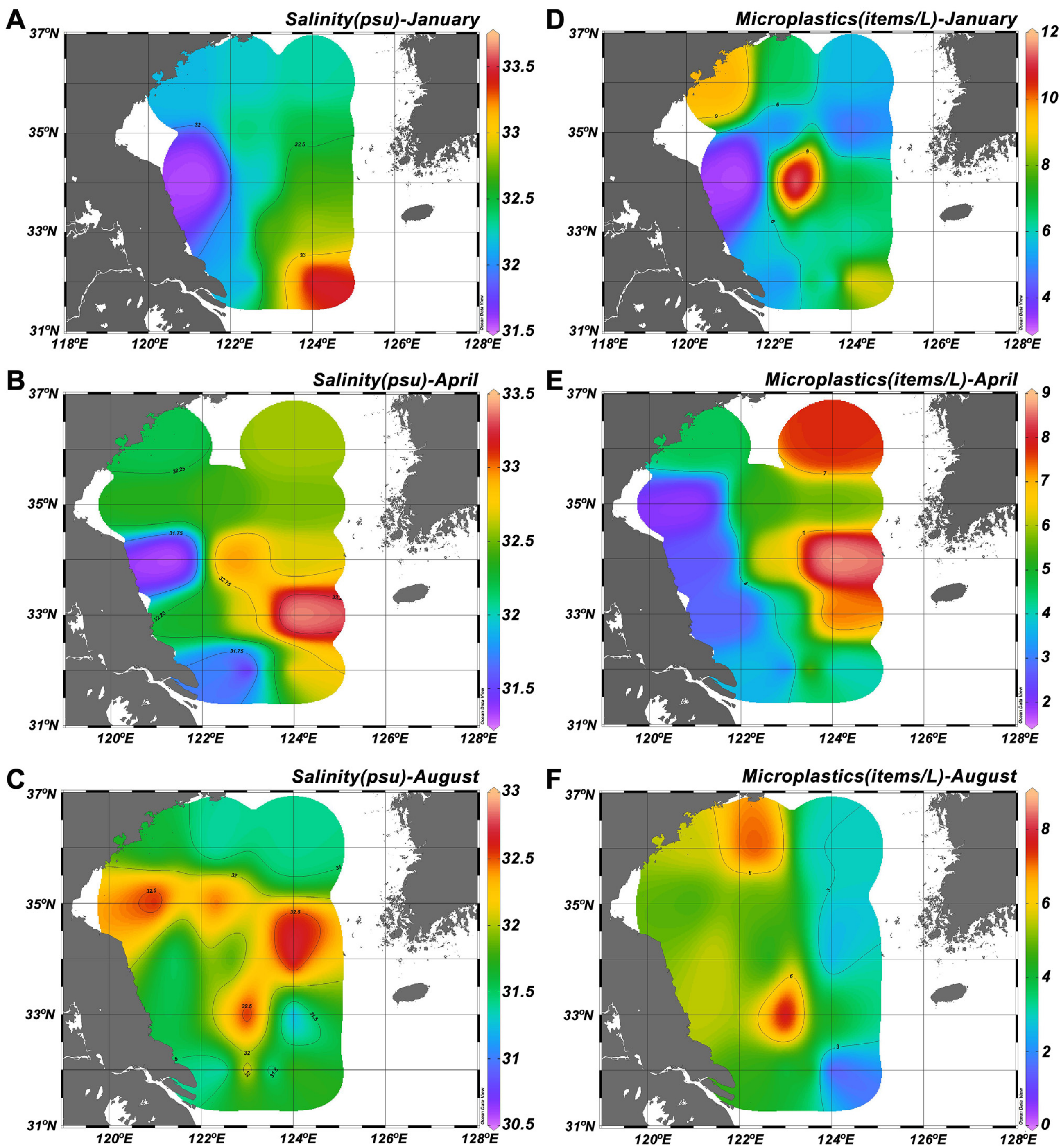


**Fig. 6.** Contour maps of salinity and microplastic abundances in the South Yellow Sea in January, April, and August of 2018.

textiles, containers, drinking straws, and automotive components (Auta et al., 2018). PA is widely used in the fishing industry (Pruter, 1987), while PET is a thermoplastic used in packaging, electronics, water bottles, and other applications (Inuwa et al., 2014). Moreover, PET and PP were found to be the major microplastic polymer types in the Yangtze River and the Hanjiang River in Wuhan (Wang et al., 2017) and in the Xiangxi Bay of the Three Gorges Reservoir (Zhang et al., 2017). Hurley et al. (Hurley et al., 2018) found that flooding could flush microplastic contamination into river catchments. Thus, it could be speculated that the increase in other microplastic types during the rainy season partly resulted from river microplastic emissions.

The spatiotemporal distribution of microplastics in the marine environment is a complex issue that is affected by several factors, including ocean hydrology, weather, physical and chemical properties of microplastics, as well as biological processes, which make it difficult to predict the transportation of marine microplastics (Fazey and Ryan, 2016; Khatmullina and Chubarenko, 2019). Even though more microplastics enter the ocean during the rainy reason, microplastic pollution in surrounding sea areas might not have increased because of the above factors. In this study, an exploration of the relationship between microplastic abundance and environmental parameters was attempted, and a positive correlation was observed between microplastic abundance and seawater salinity, indicating that salinity plays an important role in the distribution of microplastics in seawater. Different plastic materials have different densities that usually range between 0.04 and 1.58 g/cm$^3$ (Table S4, (Hidalgo-Ruz et al., 2012)). A higher salinity results in a higher buoyancy force, and thus more microplastics will float on the surface waters. In addition, the Yellow Sea Warm Current (YSWC), which is a branch of the Kuroshio Current that flows from the south to the Northern Yellow Sea, is the only exogenous oceanic water flowing into the Yellow Sea in early winter and has a relatively higher salinity and temperature (Liu et al., 2015). It is widely accepted that the YSWC is relatively strong in winter and very week or even vanishes in spring and summer (Xu et al., 2009). Thus, the YSWC flowing into the South Yellow Sea might carry along floating microplastics in winter. Additionally, the winter monsoon causes a basin-wide cyclonic surface circulation in the South Yellow Sea (Su, 2004). Reportedly, five subtropical ocean gyres might be regions at which the possible accumulation of microplastics occurs (Lebreton et al., 2012; Maximenko et al., 2012). Thus, the high abundance of microplastics, especially of small-sized ones (0.05–0.5 mm), in the winter samples (January) might be attributed to the above hydrologic features.

## 5. Conclusion

This study described the spatio-temporal distribution and characteristics of microplastics in the seawater of the South Yellow Sea and found that the abundance of microplastics, especially small-sized microplastics (50–500 μm), was relatively higher in the dry season. However, a higher variability in microplastic characteristics and composition was observed in the rainy season owing to microplastic input from land. These findings suggest that microplastic pollution in surface seawater is highly variable with respect to seasons, and this might be attributed to terrestrial sources and marine hydrological dynamics. Therefore, seasonal surveys are recommended for the accurate assessment of microplastic distribution in the ocean.

## CRediT authorship contribution statement

**Yong Jiang:**Conceptualization, Visualization, Writing - original draft. **Yanan Zhao:**Methodology, Investigation, Formal analysis.**Xin Wang:** Methodology, Validation.**Fan Yang:**Software, Data curation.**Mei Chen:** Investigation.**Jun Wang:**Supervision, Project administration, Writing - review & editing.

## Declaration of competing interest

We declare that we have no financial and personal relationships with other people or organizations that can inappropriately influence our work, there is no professional or other personal interest of any nature or kind in any product, service and/or company that could be construed as influencing the position presented in, or the review of, the manuscript entitled "Surface runoff and ocean dynamics jointly affect spatio-temporal characteristics of microplastics in the surface seawater of South Yellow Sea".

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.138375.

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