Occurrences, Distribution and Composition of Microplastics in the Surface Seawater and Beach Sediments of the Southern Shandong Peninsula, China

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Abstract

The current work provides the detailed analysis about microplastics contamination of the beach sediments and surface seawater. Sediment samples were collected from the high tidal zone at 13 beaches, and surface seawaters of seven stations along the southern Shandong Peninsula. The presence of microplastics was recorded on both beaches and surface seawater. The mean quantity of microplastics was approximately 664 ± 80 microplastics/kg of dry weight (dw) with 0.5~0.125 mm in beach sediments, and 5.62 ± 0.4 microplastics/liter (L) of surface seawater. Predominant microplastics were fibers (>97%). While other plastic forms had lower percentage. Microplastics possessed different color with polymer composition including polypropylene, polyethylene, and polystyrene. The accumulation of microplastics on beaches is an alarming issue and provides better understanding about microplastics pollution along the coastal regions of the southern Shandong Peninsula. It would also help to establish better monitoring system for future and evaluate possible risks by microplastics due to rapid economic growth of China.

Keywords: *Microplastics, Shandong, Microscopy, Fourier Transform Infrared Spectroscopy, Sediment, Beach*

Introduction

The occurrences and distribution of microplastics (<5 mm) is an alarming issue and attracted more attention and concerns in recent years (Sarijan et al. 2021). There is no standard criteria to define microplastics but particles with size 5 mm is generally separate from larger particles (Browne et al. 2010). Microplastics are mainly classified into two types: primary and secondary microplastics. The key sources of primary microplastics are household, commodities and different industrial sectors (Rochman 2015). Secondary microplastics originate from disintegration of larger plastic particles under the influence of solar radiation, temperature, biological and physical damage (Andrady 2017). Microplastics enter into marine and beach environments due to mismanagement of waste material involving marine-related activities such as fishing, recreation, swimming etc (Pereao et al. 2020). Moreover, these particles can persist in aquatic environment for long time before their mineralization after discharge (Law and Thompson 2014). Upon their entry into marine environment, various plastic materials pose threat to marine ecosystem (Anbumani et al. 2018).

Microplastics may be ingested by marine organism such as sea cucumber Stichopus horrhorrens in Pulau Pangkor (Husin et al. 2021), bacterial biofilms communities (Tavsanoglu et al. 2020), gastrointestinal tract of different species (Klangnurak et al. 2020), fish guts (Caruso et al. 2018) including human health (Sana et al. 2020). These microplastics persist in several morphologies namely fibers, spheres, films, microspheres, and fragments (Naidoo et al. 2015), with size varying from nano (n) to millimeter (mm) (Ter Halle et al. 2016). Chemically, microplastics are mainly constituted polystyrene (PS), polypropylene (PP), polyvinylchloride (PVC), polyethylene (PE), polyethylene terephthalate (PET) and few others with varying densities (Engler 2012). Microplastics are transported into ocean thru streams, rivers, storm water, and sewerage system after production at land (Doyen et al. 2019). The transporting agents are tourism, recreational activities, voyages, commercial fishing, coastal tourism, and marine-related industries (oil rigs and aquaculture) are noteworthy causing shift of microplastics to marine environment (Veerasingam et al. 2016).

Microplastics have already been identified throughout the maritime environment, containing beaches (Wu et al. 2020), open ocean (Cózar et al., 2014), remote island (Ivar et al. 2009), river (Mai et al. 2021), and marine sediment (Sharma et al. 2017). The occurrences and distribution of microplastics in beach sediment denotes long-term accumulation which acts as the persistent interfacial contact between seawater and coastal area. It was investigated that sampling from the coastal beaches is an economical and cost-effective approach to determine microplastics (Hidalgo-Ruz and Thiel 2013). When compared to mesoplastics, macroplastics and microplastics cause less visual impact when they are present in the environment; however, these cause physio-chemical imprints in food web (Ajith et al. 2020). Thus, studies about distribution and accumulation of microplastics on beaches are crucial for sustainable coastal and marine environment. A number of studies about distribution and quantification of microplastics have been accomplished on several beaches worldwide (Tiwari et al. 2019). Likewise, Colombian Caribbean (Barletta et al. 2019), Iran (Dehghani et al. 2017), Spanish (Masiá et al. 2019), and India (Gopinath et al. 2020). The transportation and distribution of microplastics on beaches may be affected with different factors such as winds, tides, anthropogenic activities, and water currents (Iwasaki et al. 2017).

A rapid economic growth has been witnessed in China during a recent past; however, about 8.82 million tons of plastic waste has been mismanaged in 192 coastal countries including China (Jambeck et al. 2015), and the awareness on the environmental and waste management was limited (Zhang et al. 2018). The Shandong Peninsula

is bound by Yellow and Bohai Sea. Numerious studies have been carried out to address microplastics issue in China (Xu et al. 2020; Shen et al. 2021; Wang et al. 2021; Yuan et al. 2021). Recently, we found few studies about the different aspects of microplastics which encompass the Yellow Sea (Sun et al. 2018, 2019; Xiong et al. 2018; Zhou et al. 2018; Zhang et al. 2019; Zhao et al. 2019; Gao et al. 2020). This is essential to assess the level, distribution, and detection of microplastics to understand the level of pollution and enlighten the contributing factors of microplastics in the region. Thus, the sand samples collected from thirteen beach sites and surface seawater samples from seven stations, and identified types, color, shape, and chemical composition. All these beach sites are located in the Shandong Province, comprising intense, highly developed and urbanized coastlines. This information will help to emphasize the gravity of microplastics pollution and highlights the critical region with other factors for an immediate attention. The work offers a better understanding regarding microplastics distribution in the coastal region of the southern Shandong, eventually encompasses better knowledge about how to improve coastal management to protect the marine ecosystems.

Materials and methods

Study Area

Shandong is one of the important coastal provinces of China with annual economic growth from marine resources in the range of 7.15 billion dollars ($) to $17.2 during 2013−18. There are famous sandy coastlines including several beaches in the southern Shandong province including Rizhao, Qingdao, and Rongcheng region. These beaches are very popular for fishing, tourism, swimming, recreational, and aqua culture-related activities adjacent to urban area of the southern Shandong Province. Tourism plays a vital role in the economy of these three cities. Especially, Qingdao receives >30 million tourists every year comprising of 88.160000.0 million foreign and domestics tourists in 2017, which were rapidly increasing on yearly basis. Qingdao is a popular tourist spot in China (Liu et al. 2019). A huge flux of beach visitors utilized these beaches for recreational and tourism activities. For example, more than 200,000 beach visitors were reckoned in the summer season on Golden bathing beach, Qingdao[1]. The present study area comprised of thirteen sandy beaches (Error! Reference source not found., E1 − E13), and seven surface seawater stations (**Error! Reference source not found.,** W1 − W7), along the southern coastline of Shandong. This area normally experiences coastal monsoons owing to its warm temperate climate and assigned as top second city in China concerning investments and efficient government (Mako 2006). The annual rainfall in this region varies from the range of 400 mm to 1335 mm, with the mean annual temperature of approximately 12.4 °C.

Sampling of sediments and water

Overall, thirteen beaches (one from Rongcheng, two from Huangdao, four from Rizhao region, and six from Qingdao region) were selected to acquire microplastics samples (**Error! Reference source not found.**). The sampling was collected in July 2019 on different dates. The detailed features of each sampling location were enlisted in supplementary information (Table S1). Number of plastics, rainfall and storm were observed during sampling period. Beach sampling techniques were selected based on the pervious standard procedures (Lots et al. 2017; Bosker et al. 2018). For each selected sampling site, a high tide line was identified to mark the boundary of

1 http://news.qingdaonews.com/qingdao/2015-08/17/content_11209692.htm (Chinese translation)

wet sand, litter, or shell deposited sites. For every beach, five sediment samples were collected around the higher tidal line along the coastline with an interval of 10~15 m with five replications for each sample. A 0.25 m^2 quadrant was employed for each beach sample. Samples were collected from the top and corner of 5 to 10 cm quadrant using a wooden ladle and five rulers. The natural debris such as leaves, seaweeds, and wood was brushed off from the samples and then transferred into aluminum boxes for further analyses. Thereafter, sediment samples were washed with distilled water in order to remove the sticky particles before sieve analysis and passed through mesh with diameter 0.038 mm in the laboratory. The particles of size ≥0.038 mm diameter was retained in the sieve. Later, samples were transferred into the Petri dish and put in oven at 60 °C for 48 hours. Then, sieve analysis test was conducted. The beach sediments of sizes 0.5~0.25, 0.25~0.125 mm was analyzed which emphasized the possible risk to marine organisms because of ingestion. The particles with size <0.125 mm was not considered in present examination because of the difficulty of sieving for further analysis. Additionally, water samples were collected in 1 L glass bottle from seven stations (W1 − W7) along different beaches of the southern Shandong Peninsula during a similar period (Error! Reference source not found.). Five samples were collected from each station with five replications for each water sample. The seawater samples were collected about 45 to 50 cm from the surface. The size of microplastics in water samples was measured by Fiji Image, J software using freehand line tool option. Moreover, the sampling glass bottles were washed with deionized water in the laboratory to delaminate impurities and airborne fibers.

Extraction of microplastics

The extraction of microplastics from beach sediments was carried out according to density separation method. This method has been frequently employed in many studies (Jayasiri et al. 2013; Kim et al. 2015). In this method, sodium chloride (NaCl) was taken as a saturated solution with an estimated density value of 1.172 g/cm^3. Here, 50 g of dried sand (sediment with grain sizes 0.5~0.25, 0.25~0.125 mm for each sample) was mixed in 250 mL saturated solution of NaCl (1.172 g/cm^3). The solution was stirred with a glass rod for about two to three minutes. Later, the solution was allowed to settle for 24 hours and finally passed through filter paper of 0.7 μm. Before analysis, the NaCl saturated solution was passed through filter paper (5 μm) to remove the additional contamination of salt crystals. All transferring devices were washed with deionized water several times and, then passed through the same filter paper to minimize loss of microplastics due to adhesion on apparatuses walls. Finally, the filter paper was sealed under a cover glass to allow drying overnight for further analysis. In case of surface seawater samples, water samples were passed through sieve of 0.5 mm mesh before laboratory examination. Thus, acquired water samples were preserved at 4 ºC till the analysis on next day (Leslie et al. 2017). Moreover, sediment and water samples were subjected to 30% H_2O_2 to digest organic matter (Tamminga et al. 2019). NaCl was added in water samples to enhance density of solution (Thompson et al. 2004). Here, 5.0 g of NaCl was added for 50 mL of each sample. Thereafter, 1 L of water sample was passed through Whatman GF/C filter paper of 0.7 μm size and sealed with glass cover to avoid external contamination and placed in glass sampling dish for drying around 24 hours at room temperature.

Quantification and characterization of microplastics

The identification of microplastics was done by stereo-microscope (Olympus SZ61 110AL2X-2 WD38) according to manufacturer's instructions (Hidalgo-Ruz et al. 2012). The microplastics falling in the following categories

were considered as microplastics. For instance, a) Homogeneous and clear b) Evident color c) No organic structure. Overall, the filter paper was segregated into four equal parts to minimize counting error during the quantification process. Each filter paper was counted three to four times for the reliable outcomes. Moreover, the number of microplastics which were estimated via visual calculations on the filter paper was confirmed with Fiji Image j software.

Polymers identification of microplastics

A FTIR Tensor 27 (Bruker, Germany) spectroscopy identified the polymer composition of microplastics. To ensure accuracy and precision, five background pooled scans were accomplished before and after every 15 measurements. The obtained spectra were matched with different spectral libraries and self-collected spectra of prior published reports. We only spectra with 70% similarity index with reference spectra were considered. The scale of study was large and we randomly selected representative samples from beach sediments ($n = 20$) for each microplastic with 0.5~0.25 and 0.25~0.125 mm, and surface seawater samples ($n = 20$) for FTIR analysis was done based on color. Four samples for each color were randomly selected to determine the polymer composition of microplastics for beach sediment and water samples. For example, White ($n = 4$), Blue ($n = 4$), Black ($n = 4$), Red ($n = 4$), and Others ($n = 4$).

Methodology validation for microplastics color

Based on color, microplastics were categorized into five groups: white, blue, black, red, and others. The white category included colorless and transparent microplastics. The blue group contained deep blue, light blue, deep green, and light green microplastics. The black category comprised transparent black, gray, and white-striped black microplastics. The red group possessed pink and purple microplastics, while other groups consisted of different color rather than above categories.

Statistical and data analysis

Probability plot, Levine test, and Shapiro Wilk tests were performed to determine the normality of data. One-Way ANOVA and Spearman's rank correlation was used to determine the significant difference and relationship between the number of microplastics related to grain size. Whereas, Two-Way ANOVA was performed to figure out the significant difference between the color categories of microplastics at different locations for both beach sediment sites ($E1 - E13$), and surface seawater stations ($W1 - W7$).

Quality assurance (QA) and quality control (QC)

All apparatus and instruments were washed with Milli Q water before performing the experiment. The sampling and experimental devices were covered with aluminum foil during laboratory work in order to avoid contamination. Therefore, non-textile robe, gloves, non-plastic tolls, and caps were used to minimize the effect of clothes and hairs. The door and windows of laboratory were closed during experiment. The filter paper was covered with glass to protect from air contamination. Moreover, the known sample of sodium and potassium bromide were run before FTIR analysis to ensure the validity of FTIR results. Ethanol was used to clean the germanium crystal of FTIR.

Results

Abundance of microplastics in beach sediment

The findings of present study showed that every sample in beach sediments possessed microplastics. The mean microplastic abundance of thirteen sites was approximately 664 ± 80 microplastic/kg dw with 0.5~0.25 and 0.25~0.125 mm. The median of microplastics was estimated to be 236 and 380 corresponding with grain size 0.5~0.25 and 0.25~0.125 mm, respectively. The microplastics with the size of 0.25~0.125 mm contained higher concentration of microplastics than those with size of 0.5~0.25 mm. When the concentration of microplastics obtained from different beaches was compared, higher concentration of microplastics was noted for Bathing beach No.1 (E6), i.e. 716 ± 72, and 900 ± 180 microplastics/kg dw for 0.5~0.25 and 0.25~0.125 mm followed by Wanpingkou bathing beach (E13) with 396 ± 16 and 352 ± 52 microplastics/kg dw with 0.5~0.25 and 0.25~0.125 mm and so on (See Error! Reference source not found. for comparison). Whereas, a lower concentration of microplastics (172 ± 12, and 256 ± 20 microplastics/kg dw with 0.5~0.25 and 0.25~0.125 mm) was found at Chengdao beach (E1). One-Way Anova results revealed a significant difference between the number of microplastics and sediments with grain size of 0.5~0.25, 0.25~0.125 mm. Spearman's rank correlation revealed a positive relationship between microplastic abundance and grain size. The abundance of microplastics increased with the decreasing grain size. It indicated that distribution and abundance of microplastics were statistically same among different sites.

Abundance of microplastics in surface seawater

Each beach sediment and sample of surface seawater contained microplastics. The average microplastic abundance for all stations $(W1 - W7)$ was 5.6 ± 0.4 microplastics/L. A variation of microplastics was found at each station. The abundance and distribution of microplastics in surface seawater at different stations was presented in Error! Reference source not found.**.** Moreover, tolls of Fiji image J software were utilized to determine the size of microplastics for surface seawater. Microplastics sizes < 125 μm had higher concentrations followed by 125~250 and 250~500 μm (**Error! Reference source not found.**). Statistically significant difference was found between microplastic abundance and grain size.

Color of Microplastics

The variations in the colors of microplastics were observed in both surface seawater and beach sediments which were assigned five categories as discussed in the methodology section. The color of microplastics varied in beach sediments and surface seawater. Out of five categories, black microplastics were dominant (approximately 34%) followed by white, blue, red and others accounted as 22%, 22%, 16% and 6% of beach sediment with 0.5~0.25 mm for all sites. Similarly, sediment with 0.25~0.125 mm contained mostly white microplastics (37%) followed by back, blue, red, and others represented by 28%, 16%, 13%, and 6%. Besides, the highest proportion of white microplastics was observed at E9 (48%) and the lowest in E8 (21%). Additionally, greater proportion of black microplastics was estimated to be 52% at W4 and the lowest in W3 accounting 28%. Overall, black microplastics were dominant (40%), followed by blue, white, red and others comprising 24%, 17%, 13%, and 6% (**Error! Reference source not found.**).

Shape of Microplastics

Fibers were the dominant shape in both sediment and seawater. These fibers possessed multiple colors as discussed in the previous section. Out of 100%, average microplastic abundance, approximately 97% were fibers, and remaining were others including fragments in beach sediments at different sites which was similar to surface seawater (Error! Reference source not found.). Moreover, no statistical difference was noticed within the fibers and other microplastics for both beach and seawater at different stations.

Polymer identification of microplastics

Numerous spectral peaks attained by FTIR spectroscopy which were in the range of 400~4000 cm^{-1} due to the presence of microplastics. These spectral peaks attributed to different functional groups. The different spectral peaks on various wavelengths demonstrated the existence of multiple features such as vibrations, stretching, bounding, and aromatic rings containing different elements. The most common microplastic polymers included polypropylene (PP), polyethylene (PE), polystyrene (PS) with density ranges of 0.895~0.92, 0.93~0.97, and 0.96~1.05 g/cm^3) in beach sediments and surface seawater. The densities of these polymers were similar to the density of water (Hidalgo-Ruz et al., 2012). In **Error! Reference source not found.**a, sharp peaks were observed at 2915 cm^{-1}, 2848 cm^{-1} and 1465 cm^{-1} belong with $–CH_2–$ vibration. The peak at 721 cm^{-1} attributed to $–CH_2–$ in-plane oscillating vibration. Therefore, acquired spectra were PE (**Error! Reference source not found.**a). Similarly, **Error! Reference source not found.**b comprised of three peaks at 1377 cm^{-1}, 1156 cm^{-1} and 971 cm^{-1} due to $–CH_3$ and $–CH_2–$ vibration with peaks at 2953 cm^{-1}, 2917 cm^{-1}, 2845 cm^{-1} and 1459 cm^{-1}. Thus, the obtained spectra were PP (**Error! Reference source not found.**b). Furthermore, **Error! Reference source not found.**c showed the spectra of PS though several sharp peaks developed at different absorbance bands. For example, sharp peaks at 3024 cm^{-1}, and 2847 cm^{-1} showed C–H stretches, and 1601 cm^{-1}, 1492 cm^{-1} attributed to the aromatic rings, and 1041 cm^{-1}, 1027 cm^{-1} and 694 cm^{-1} were because of $–CH_2–$ and aromatic CH bend. The absorption of determining polymers was identified via spectral libraries and direct matching of prior attained spectra (Verleye et al. 2001; Nishikida and Coates 2003; Noda et al. 2007; Asensio et al. 2009). PS was more abundant polymer than PP and PE in both beach and water samples. The percentage of PS was estimated to be 50%, 40% beach sites with 0.5~0.25, 0.25~0.125 mm, and 40% in seawater followed by PP and PE. The proportion of these polymers was based on color as shown in **Error! Reference source not found.**.

Discussion

The concentration of microplastics was found ubiquitous on designated beaches and surface seawaters in the present study. The higher concentration of microplastics in beach sediments than surface seawater might be due to regular and long-term accumulation via ocean currents including terrestrial origin. Whereas, the lesser concentration of microplastics in surface seawater could be attributed to numerous factors such as hydrodynamics, sample handling method, ocean gyres, tides, rainfall, wind and sampling season etc. (Kukulka et al. 2012) documented several factors that could influence the abundance and distribution of microplastics. These factors were wind (Collignon et al. 2012), storm (Lattin et al. 2004), shape, size, density, and settling velocity of microplastics in the water column (Khatmullina and Isachenko 2017). Moreover, there is no specified criterion and methodology for the collection, quantification, and characterization of microplastics (Besley et al. 2017). It was not easy to outline relationship between microplastic abundance with previous studies elsewhere. The findings

of present study were compared with other studies which applied similar units for quantification of microplastics for beach sediments (microplastics/kg dw), and surface water (microplastics/L). The microplastic abundance along the beaches of southern Shandong was not uniform in comparison to the previous studies (Error! Reference source not found.). For instance, the mean microplastics on the southern Shandong beaches was 664 ± 80 microplastics/kg dw, compared with 261 microplastics/kg dw on the Caribbean beaches (Bosker et al. 2018) and 248 microplastics/kg dw on the European beaches (Lots et al. 2017). These coastlines contained lesser concentration of microplastics than the southern Shandong beaches. Whereas, Halifax Harbor, Canada, Venice Lagoon, and Puducherry Coast was contaminated with higher microplastics concentration i.e. $4500, 720.30 \pm 191.60, 1454$ microplastics/kg dw. Out of designated beaches, the Bathing beach No.1 (E6) contained higher concentration of microplastics i.e. 716 ± 72 microplastics/kg dw, 900 ± 180 microplastics/kg dw with 0.5~0.25, and 0.25~0.125 mm than other beaches. Overall, variation of microplastics at different sites whether globally or locally seemed due to numerous reasons. The concentrations of microplastics were linked with population density and beachgoers (Pedrotti et al. 2016). Low concentration of microplastics could be due to lesser anthropological activities (Nor and Obbard 2014), though these activities were the primary source of micro-pollutants distribution (Vianello et al. 2013), including industrialization, urbanization, sampling procedure, sampling season with natural factors like, wind, rainfall, storms and geographic proximity of the region. For example, the concentration of microplastics was significantly greater on windward beaches than leeward beaches (Monteiro et al. 2018). For the present study, the higher microplastics concentration at Bathing beach No.1 (E6) might be due to immense swimming, shipping, fishing, and beachgoers activities with some other factors during sampling period. Moreover, the determination of the absolute information about variations of microplastics on the beaches of Shandong Peninsula than other beaches elsewhere was challenging task due to diverse anthropogenic and natural factors.

The average microplastic abundance of sea water was estimated to be 5.6 ± 0.4 microplastics/L which was moderate than many studies elsewhere (Error! Reference source not found.). The variation of microplastics distribution in maritime environment involved multiple factors comprising weather, hydrology, physiochemical, and biological processes that make it difficult to predict mode of microplastics in marine environment (Khatmullina and Chubarenko 2019). In addition, the higher salinity enhances buoyancy which supports floating of microplastics on surface of seawater. The Yellow Sea encompass numerous currents (Liu et al. 2015), which vary in different seasons (Xu et al. 2018). These currents could distribute different materials including microplastics (Jilan 2004). Thus, it has been hypothesized that difference in microplastic abundance in the present study might be different due to multiple hydrological features than elsewhere. The presence of microplastics in surface seawater of the Yellow Sea near coastal regions of the southern Shandong showed that, microplastics accumulation on different beaches occurred via ocean currents and tides. Moreover, beachgoers, urban run-off, sewage discharge, recreational, shipping, swimming activities, industrialization, and urbanization have significant role in variability of microplastics. Moreover, we also found few recent studies about different aspects of microplastics that encompass the Yellow Sea. Likewise, characterization and distribution of microplastics (Jiang et al. 2020b), investigation of microplastics in the digestive tract of marine species (Sun et al. 2019), zooplankton (Sun et al. 2018), prevalence of microplastics in sediment and benthic organism (Guo and Wang 2019), offshore sediment (Zhang et al. 2018).

Importantly, the present study showed that the majority of microplastics was fibers and very few were others in beach sediments and surface seawater. Our findings were underlined with prior outcomes of Caribbean, European, and Mexico beaches (**Error! Reference source not found.**). Those studies found fibers to have higher percentage (>90%) of microplastics (Lots et al. 2017; Bosker et al. 2018). Immense human activities produce microfibers during washing, production, and natural aging in textile industries. A huge amount of fibers enters into marine environment along sewage discharge without proper treatment, high development and industrialization (Zhao et al. 2018). The microplastics were noticed multicolor in this study which showed different sources of microplastics. The color of microplastics has been used to determine their origin in different environments (Li et al. 2018). For example, microplastics with blue, violet, red, or green color were commonly made of polyester and acrylic (de Jesus Piñon-Colin et al. 2018). Similarly, white microplastics were composed of PP though translucent microplastics were mainly composed of PE on La Graciosa Island, Spain (Edo et al. 2019). Microplastics color is a crucial feature assessing the adsorption relationship and interaction with couple of species and was intended that dark black microplastics with multicolored microplastics had greater chances of ingestion as food by mistake (Ory et al. 2017). The presence of different microplastics colors revealed the possible hazard and risk to marine species along the southern Shandong Peninsula. Moreover, it could directly affect human health through the consumption of different marine species.

FTIR analysis of this study showed the prevalence of PE, PS, and PP in surface seawater and beach sediments. These polymers were recognized more prominently in different coastal and marine environments (Simon-Sánchez et al. 2019). The large quantity of these polymers in coastal and marine environments was due to immense aquaculture, agriculture, and rapid industrial developments (Facts 2019). Out of these polymers, PE was more common and utilized in food packing, agricultural films, plastic bags, ties, clothes, ropes, carpets, and bottles. While, PP was an integral part of food packing, pipes, carpets, straws, water bottle caps, and plastic containers. PS was primarily used in plates, disposable cups, textile industry, and Styrofoam material. The existence of higher percentage of these polymers (PE, PP, and PS) in maritime environment might be due to higher output which possessed less density than water and float on the surface water (Europe 2015). These polymers were also being produced on the account of global plastic production and discharge into the sea without proper treatment involving local sources such as local industrial waste, urbanization, disposal of sewage waste, fishing and shipping etc (Lots et al. 2017). The polymers on the southern Shandong beaches could be transported via ocean currents or tides and then accumulated with the process of sedimentation.

In last, the findings of this study will assist to develop a better understanding of microplastics occurrence including source and pathways in marine and coastal zones of the southern Shandong Peninsula. It also facilitates establishing better monitoring system and provides beneficial data to evaluate the possible risk to the coastal zone and marine ecosystems caused by microplastics. A detailed investigation on microplastics would be beneficent regarding marine food-related species including fishes along the southern part of the Yellow Sea to assess possible risk to human health due to the consumption of microplastics via seafood. Moreover, the effect of microplastics on the properties of these beaches is yet unknown. We highly recommend the utilization of different simulation models to conduct field surveys for sake of measuring an actual magnitude of plastic pollution at a global scale considering biological processes, coupled with physical-biological models for marine microplastics. The sampling and analysis method should be globally standardized for better comparison of the microplastic abundance among different regions. We used method which only detect the microplastics with density < 1.17 g/cm^3. Therefore, we

were unable to determine the polymers with high density such as polyethylene terephthalate, rayon, polyvinyl chloride, etc. (density 1.39, 1.56, and 1.53 g per cm^3) and so many others due to adaptation of method but we were still capable to segregate the most common polymers which have been identified worldwide.

Conclusion

Identification, characterization, and quantification of microplastics were analyzed on thirteen beaches of the southern Shandong and seven stations in the surface seawater of the southern Yellow Sea. The mean abundance of microplastics was approximately 664 ± 80 microplastics/kg dw in beach sediment and 5.62 ± 0.49 microplastics/L in surface seawater. The concentration of microplastics increased with decrease of their grain sizes. The prominent morphology of microplastics was fibers with few fragments. The color of microplastics for both sediments and surface seawater was mainly white, black, blue, and red constituting PP, PE, and PS polymer composition. The source of microplastics on the beaches might be ocean currents or tides and some terrestrial origin. Whereas, global and local sources such as disposal of industrial wastes, sewage waste etc. without proper treatment could be other source of microplastics to the Yellow Sea. This study provides comprehensive and novel data on the microplastics distribution on different beaches of the southern Shandong Peninsula that allude to proper mitigation and monitoring plans to overcome microplastics risk of the coastal and marine ecosystems. Otherwise, the number of microplastics would keep on increasing dramatically which would have severe consequences to marine species including human health.

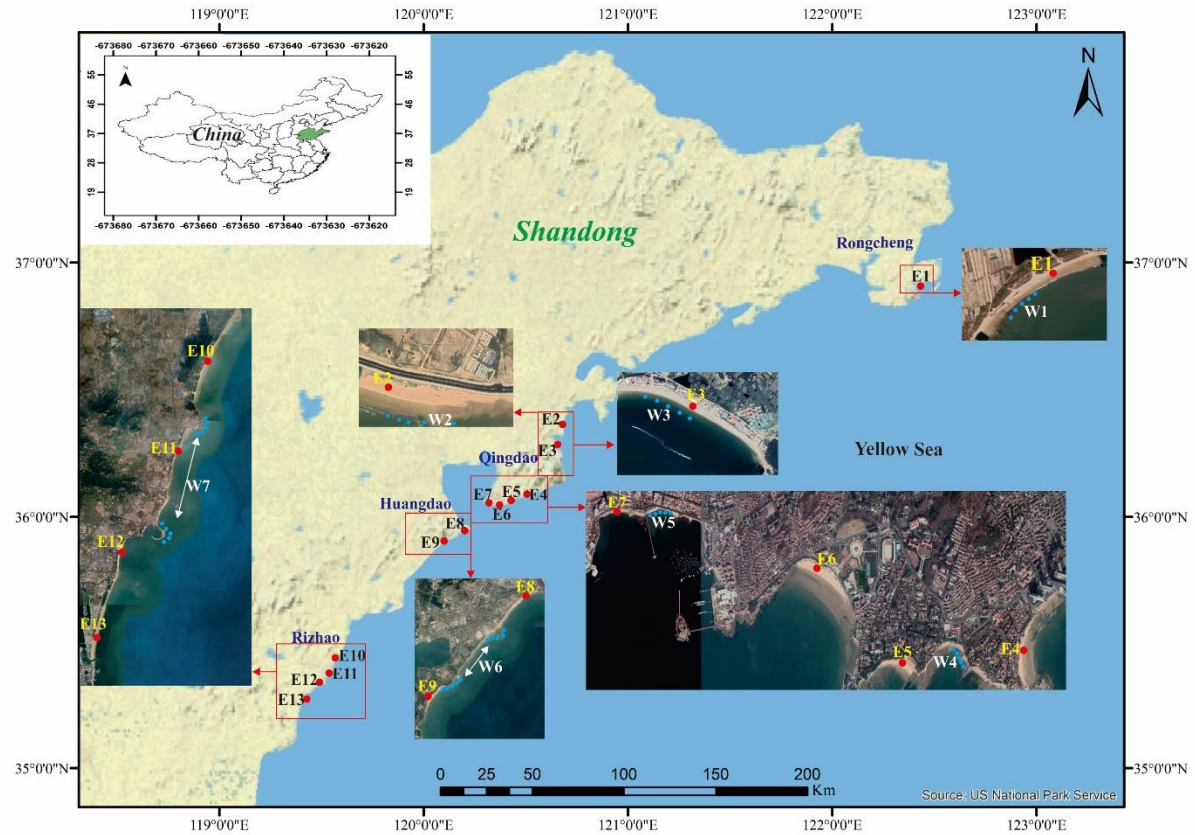


Fig 1 Map of the study area. $W1 - W7$ represent then surface seawater stations, and $E1 - E13$ denotes the beach sites.

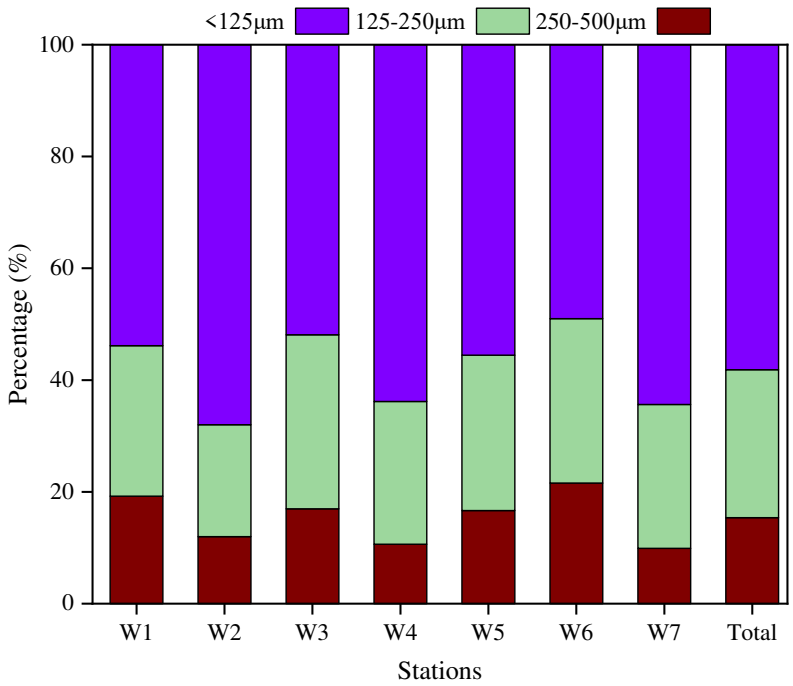


Fig 2 The microplastic abundance and sizes at various stations. The size of microplastics for surface seawater samples was determined by Fiji Image J software.

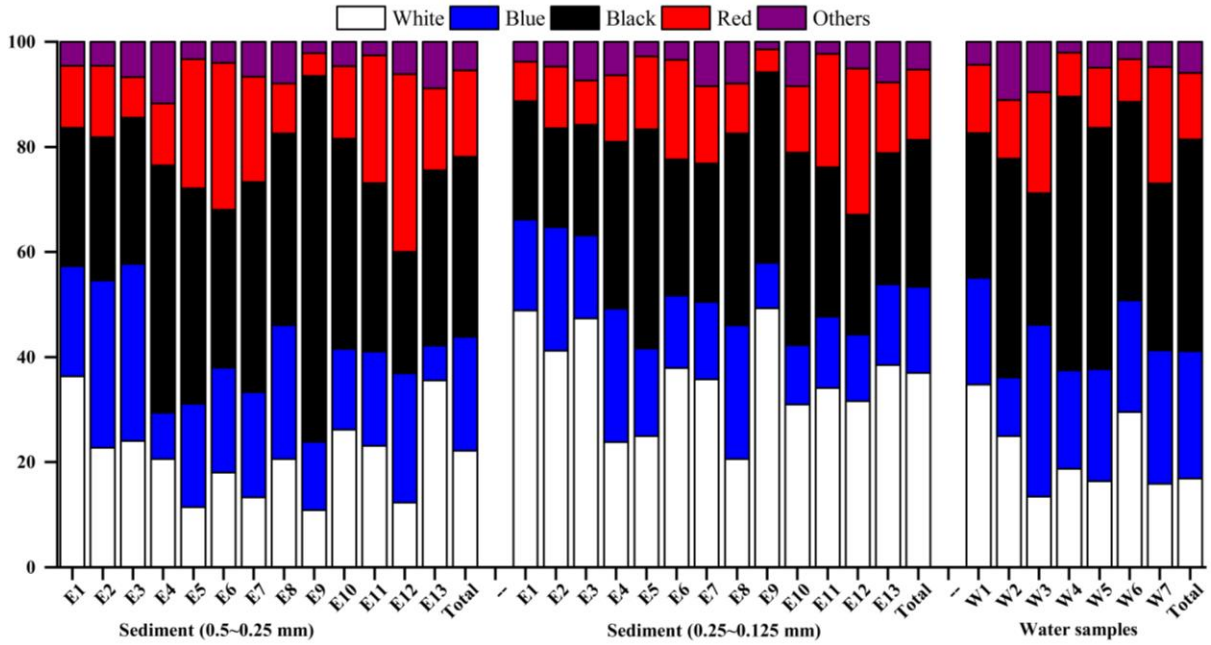


Fig 3 The proportion of different color of beach sediments and water samples at different sites.

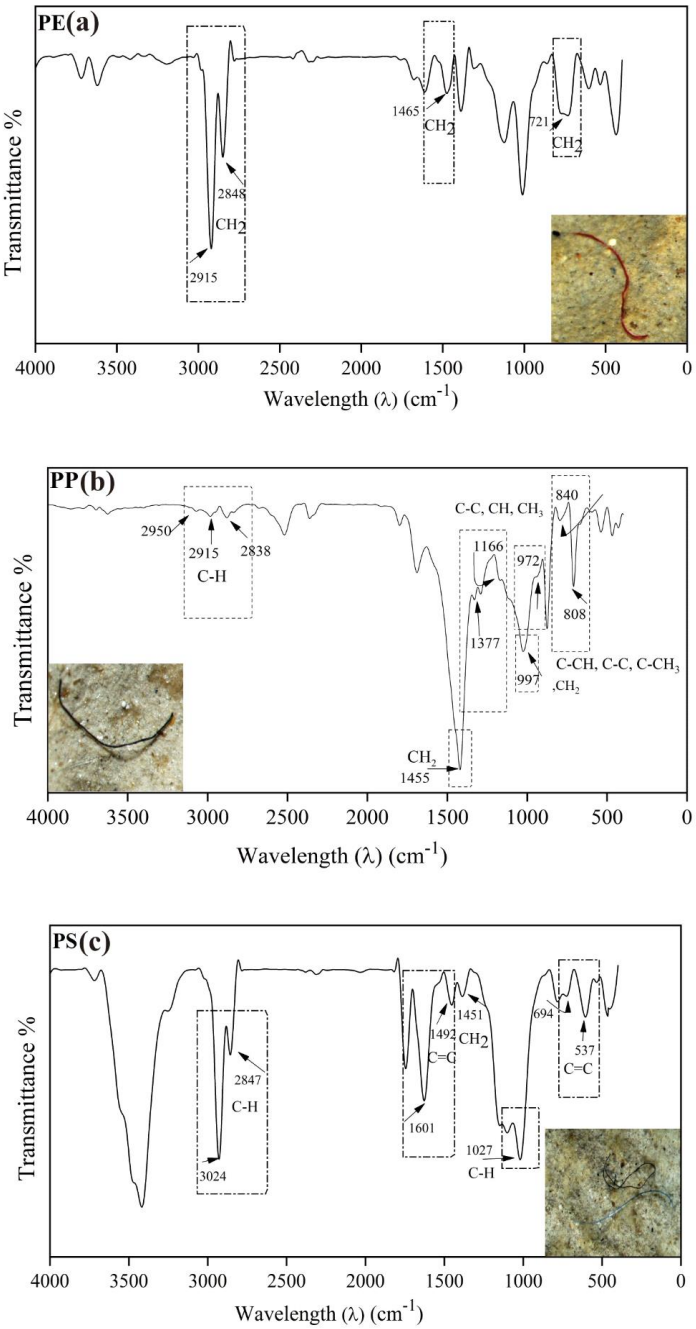
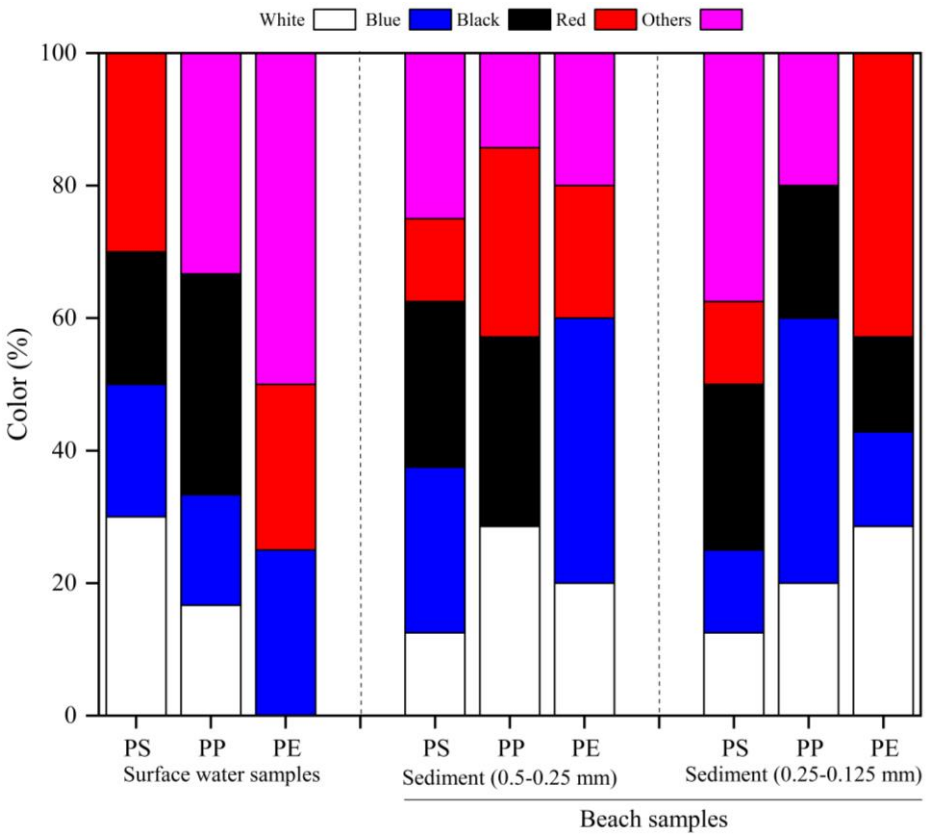


Fig 4 Illustration of the spectra of microplastics polymers including peaks of (polyethylene (PE), polypropylene (PP), polystyrene (PS)) from both surface seawater and beach sediments via FTIR spectroscopy.



342 *Fig 5 The proportion of different polymers with respect to color for beach sediments and surface seawater.*

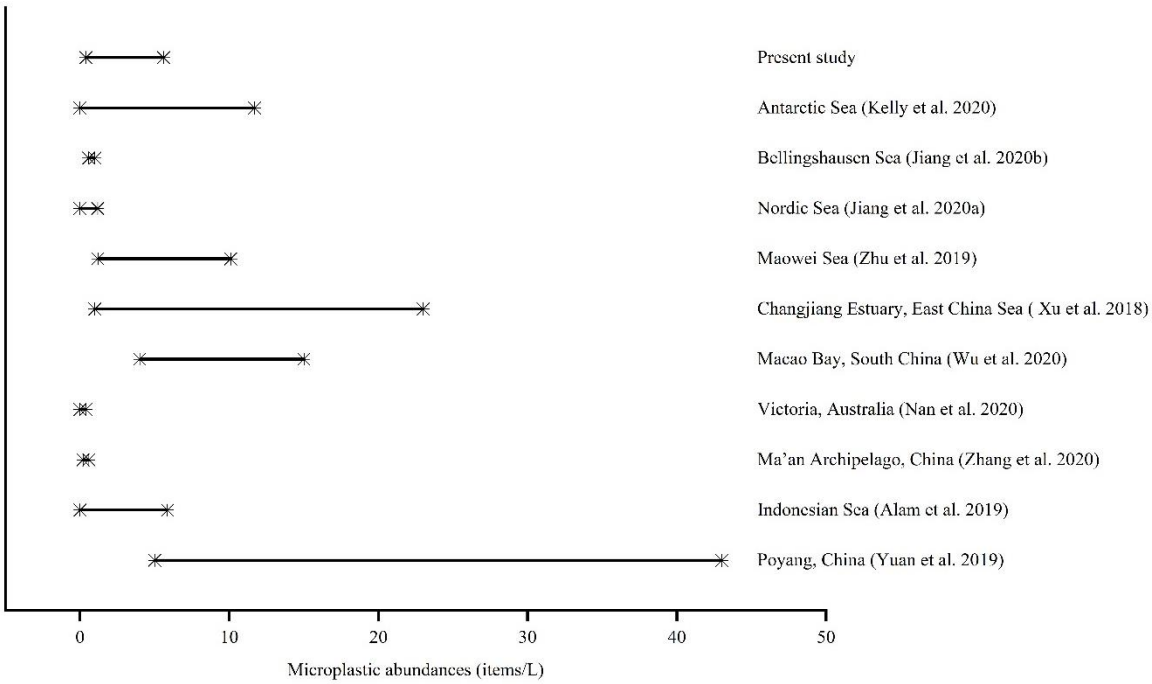


Fig 6 Microplastic abundance in different seas including present study.

343

Table 1 Microplastic abundance and type of beach sediments with different grain sizes. Results are presented from different sites and assigned (E1−E13) to represent each site. Microplastic abundance of each site was taken as an average number of microplastics from 5 replications per kg of the dry sediment (dw) (± SEM). Microplastics (Mps) are classified between fibers and others and expressed in proportion of total count.

Location	Sediment grain size (mm)	Mps/ kg dw	Mps Type	
			Fibers (%)	Others (%)
E1 (Chengdao beach)	0.5~0.25	172 ± 12	96	4
	0.25~0.125	256 ± 20	97	3
E2 (Aoshan beach)	0.5~0.25	180 ± 12	97	3
	0.25~0.125	352 ± 16	97	3
E3 (Yangkou beach)	0.5~0.25	252 ± 24	97	3
	0.25~0.125	380 ± 60	98	2
E4 (Bathing beach 3)	0.5~0.25	200 ± 12	97	3
	0.25~0.125	300 ± 32	98	2
E5 (Bathing beach 2)	0.5~0.25	192 ± 16	98	2
	0.25~0.125	388 ± 28	98	2
E6 (Bathing beach No.1)	0.5~0.25	716 ± 72	96	4
	0.25~0.125	900 ± 180	97	3
E7 (Zhanqiao beach)	0.5~0.25	240 ± 28	97	3
	0.25~0.125	368 ± 72	97	3
E8 (Golden bathing beach)	0.5~0.25	244 ± 20	97	3
	0.25~0.125	440 ± 48	97	3
E9 (Silver beach)	0.5~0.25	260 ± 20	97	3
	0.25~0.125	420 ± 140	97	3
E10 (Forest park bathing beach)	0.5~0.25	224 ± 12	96	4
	0.25~0.125	396 ± 40	98	2
E11 (Dachenjia beach)	0.5~0.25	212 ± 8	97	3
	0.25~0.125	260 ± 24	98	2
E12 (Dongxiaozhuang beach)	0.5~0.25	236 ± 16	96	4
	0.25~0.125	404 ± 36	97	3
E13 (Wanpingkou bathing beach)	0.5~0.25	396 ± 16	96	4
	0.25~0.125	352 ± 52	97	3
Overall Mps mean		664 ± 80	97	3

348 *Table 2 The microplastic abundance was taken as total mean of each station for (N) number of samples with 5*
349 *replications and expressed in mean (L^{-1}) with standard error mean (SEM) of surface seawater. A symbol* $(W1 - W7)$ *is given to represent each station.*
350

Region Name	N	Microplastics mean (L^{-1})	% Fibers	% Others
W1 (Chengdao beach)	5	4 ± 0.4	97	3
W2 (Aoshan beach)	5	4.6 ± 0.6	98	2
W3 (Yangkou beach)	5	5.4 ± 0.6	97	3
W4 (Bathing beach 2 & 3)	10	5 ± 0.5	97	3
W5 (Bathing No.1 & Zhanqiao beach)	10	9.2 ± 0.4	97	3
W6 (Golden & Silver beach)	10	6 ± 0.4	98	2
W7 (Rizhao coastal area)	10	5.2 ± 0.2	96	4
Overall, mean value	55	5.6 ± 0.4	97	3

351 *Table 3 Mean grain size of microplastics contents along different beaches worldwide (Particles per kg dw).*

Location	Sediment size (mm)	Mean	Median	Sources
Hongkong	2~0.063	16.8	5	(Lo et al. 2018)
Belgium	1~0.038	134	-	(Claessens et al. 2011)
Beibu Gulf, China	< 0.5	6870	6140	(Qiu et al. 2015)
Caribbean	< 0.5	261 ± 6	276	(Bosker et al. 2018)
European	< 1	248	143	(Lots et al. 2017)
Persian Gulf	1~0.01	61 ± 49	-	(Naji et al. 2017)
Halifax Harbor, Canada	0.5~0.063	4500	3950	(Mathalon and Hill 2014)
Isle of Rügen, Germany	> 0.5	93.0	88.1	(Hengstmann et al. 2018)
Puducherry Coast	1~0.3	720 ± 192	-	(Dowarah and Devipriya 2019)
Qatar	1~5	$6 - 38$ (13.5)	-	(Abayomi et al. 2017)
Venice Lagoon, Italy	< 1	1445	1435	(Vianello et al. 2013)
Shandong, China	**0.5~0.125**	664 ± 80	**308**	**Present study**

352

353 *Table 4 Prior investigations about microplastics concerning different characteristics worldwide.*

Country	Types of microplastics	Color	Chemical composition	Reference
Brazil	Fragments (56 %), Foam (26.7 %), Pellet (9.9 %)	Fibers: blue, red and green. Pellets: translucent	Fibers PP, Nylon & Polyvinyl alcohol. PS (26.7 %)	(de Carvalho and Neto 2016)
Colombia	Pellet	White (65.07 %), other, (24.26 %), gray (3.83 %)	PE and PP	(Acosta-Coley and Olivero-Verbel 2015)
Europe	Fibers (98.7 %), Particles (0.91 %), Film (0.35 %)	Blue/black (77.5~82.9 %), red (9.3~13.6%)	Polyester (70 %), PP (20 %), PE (10 %)	(Lots et al. 2017)
India	Fragments (47–50 %), Fibers (24~27 %) and Foam (10~19 %)	N. R	PE (45.98 %), PP (19.41 %), PS (17.41 %)	(Karthik et al. 2018)
Mexico	Fibers (91 %), Film (5 %), Spheres (toys) (3 %)	Fibers: black (59 %) and blue (25%). Others: (12%)	Polyacrylamide, Nylon, Polyacrylate	(de Jesus Piñon-Colin et al. 2018)
Mexico	Foams (15 %), Fibers (11 %), Rigid and semirigid fragments (56 %), Film fragments (10 %), Pellets and ammunition for toys (8 %)	White (23 %), green (17%), blue (17%), yellow (16%), transparent (11%), red (6%), others (10%)	PE (56 %), PP (21 %), PS (12 %), others (11 %)	(Alvarez-Zeferino et al. 2020)
Caribbean	Fibers (> 95 %), Others (5 %)	N. R	N. R	(Bosker et al. 2018)
Portugal	Fibers (80.64 %) and Fragments (19.63%)	Fibers: red, green, blue and black.	Fibers: Rayon (81 %). Fragments: PP (19.36 %)	(Frias et al. 2016, p. 20)
Shandong	**Fibers (98%), and Fragments (2%)**	**Transparent, blue, red, black, and others**	**PP, PE and PS**	**Present study**

354 N. R= Not reported; PE=polyethylene, PP=polypropylene, PS=polystyren

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Author contributions

Rashid Pervez: Conceptualization, Formal analysis, Investigation, Writing - review & editing. Yonghong Wang: Supervision, ZhongPeng Lai: English Editing and proof reading.

Data availability

The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Ethical approval

Not applicable

Consent to participate

Informed consent was obtained from all individual participants included in the study.

Consent to participate

Not applicable

Competing interest

The authors declare no conflict of interest.

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