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Articles

A novel in situ and graded sampling system for seawater microplastics

Un novedoso sistema de muestreo *in situ* y de clasificación para microplásticos presentes en el agua del mar

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Abstract

The net trawling method requires complicated operations, during which samples are susceptible to contamination in an open environment. In this context, a novel *in situ* and graded sampling system for seawater microplastics was developed. This system integrates the collection, separation, and enrichment procedures and features alternate air-water elution and three-layer graded collection (membrane mesh sizes of 300, 100, and 50 μm). Its advantages are ease of operation, reduced operation time, seawater recycling, and forming a closed space to avoid sample contamination. Moreover, it is capable of measuring the filtered seawater volume accurately. An in-shore experiment was carried out to collect microplastic samples with the new system. These samples were analyzed and identified by micro-Raman spectroscopy. Microplastics with different types (PE, PET, PA, PEVA, PS, and PVS); colors (translucent, white, blue, and yellowish), and shapes (foam, fragment, fiber, and granule) were found on the 100 and 50 μm filter membranes but not on the 300 μm filter membrane. The results indicate that the new sampling system effectively collects microplastics usually omitted by trawling nets and can be used as a supplement to trawling nets.

Keywords: Microplastics, seawater microplastics, sampling system, *in situ*.

Resumen

Como nuevo tipo de contaminante, los microplásticos han estado atrayendo una gran atención durante el proceso de monitorización del medio marino. Las redes de arrastre utilizadas en la actualidad (con un tamaño de malla de 330 µm) no son realmente adecuadas para la recogida de partículas de microplástico más pequeñas debido a su gran tamaño de malla. Además, el método de captura con redes de arrastre requiere de operaciones complicadas, durante las cuales las muestras son susceptibles de contaminación en un entorno abierto. En este contexto, se desarrolló un novedoso sistema de muestreo *in situ* y de clasificación de microplásticos de agua de mar que integra un procedimiento de recogida, separación y enriquecimiento, y cuenta también con un sistema de elución alternativo aire-agua y de recogida con clasificación de tres capas (con tamaños de malla de membrana de 300, 100 y 50 µm). Entre sus ventajas se incluyen su facilidad de utilización, la reducción en el tiempo de su operación, el reciclado del agua del mar y la formación de un espacio cerrado con el fin de evitar la contaminación de las muestras. Además, es capaz de medir con precisión el volumen de agua de mar filtrada y las pruebas de elución realizadas con él demuestran que su tasa de recuperación alcanza más de un 90 % en cinco segundos. Posteriormente se llevó a cabo un experimento en tierra con el fin de recoger muestras de microplásticos utilizando el nuevo sistema. Las muestras después fueron analizadas e identificadas por espectroscopia micro-Raman, encontrándose microplásticos de diferentes tipos (PE, PET, PA, PEVA, PS y PVS); colores (translúcido, blanco, azul y amarillo), y formas (espuma, fragmentos, fibras y gránulos) en las membranas filtrantes de 100 y 50 μm, pero no en la membrana filtrante de 300 μm. Los resultados del estudio revelan que el nuevo sistema de muestreo resulta realmente eficaz en la recogida de microplásticos que por lo regular es omitida por las redes de arrastre y que puede utilizarse como complemento a dicho sistema.

Palabras clave: microplásticos, microplásticos de agua de mar, sistema de muestreo, *in situ*.

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Introduction

Microplastics, defined as plastic particles smaller than 5.0 mm (Arthur, Baker, & Bamford, 2008), have stable chemical properties and can exist in seawater for a long time (Wang, Lin, & Yuan, 2017). Microplastics have been attracting attention in marine environmental monitoring as a new type of pollutant. Although pollution by seawater microplastics was noticed in the early 1970s (Carpenter & Smith, 1972; Carpenter, Anderson, Harvey, Miklas, & Peck, 1972; Wong, Green, & Cretney, 1974), the term "microplastics" was proposed in 2004 for the first time (Thompson, Olsen, Mitchell, Davis, Rowland, John, McGonigle, & Russell, 2004). Since then, scientific communities have begun to pay full attention to the possible ecological and environmental problems caused by these fine plastic particles in the ocean. There are two main types of sources of microplastics: the "primary" microplastic sources are those in which microplastics are intentionally produced either for direct use or as precursors to other products, and the "secondary" microplastics are formed in the environment from the breakdown of larger plastic material, especially marine Debris. Due to their small size, microplastics have the potential to be ingested by benthic and planktonic organisms. Microplastics with a very low biodegradation potential entering marine food webs will endanger the safety of marine life. Scientific studies have confirmed that seawater microplastics impact growth and development (Goldstein, Rosenberg, & Cheng, 2012; Rochman, Tahir, Williams, Baxa, Lam, Miller, Teh, Werorilangi, & Teh, 2015; Lu, Zhang, Deng, Jiang, Zhao, Geng, Ding, & Ren, 2016; Zhang, Chen, Wang, & Tan, 2017a; Lo & Chan, 2018) and reproduction of marine organisms (Browne, Dissanayake, Galloway, Lowe, & Thompson, 2008; Law, Morét-Ferguson, Goodwin, Zettler, DeForce, Kukulka, & Proskurowski, 2014; Zhang, Wang, Song, Fang, & Wang, 2020). Thus, the detailed qualitative and quantitative monitoring of microplastics in the marine environment is highly required.

As is known, sampling and separation are the two key steps in researching seawater microplastics. Trawling nets and pumps are the most commonly used sampling equipment for microplastic collection in marine environmental monitoring. According to the working depth in seawater, trawling nets are usually divided into four categories (Rocha-Santos & Duarte, 2014): (1) manta trawls (Faure, Saini, Potter, Galgani, Alencastro, & Hagmann, 2015) (Figure 1(a)) and neuston trawls (Cózar, Sanz-Martin, Marti, González-Gordillo, Ubeda, Gálvez, Irigoien, & Duarte, 2015) (Figure 1(b)), which are usually used for surface water, (2) bongo trawls (Cole, Lindeque, Halsband, & Galloway, 2011) (Figure 1(c)), which are usually used for middle water, (3) benthic trawls, which are used for deep bottom water, and (4) containers such as buckets that are suitable for collecting large samples (Luo, Lin, Jia, Xu, & Li, 2019). Due to the lack of standardization of sampling methodologies for microplastics, published studies have adopted different types of sampling nets with varying mesh sizes.

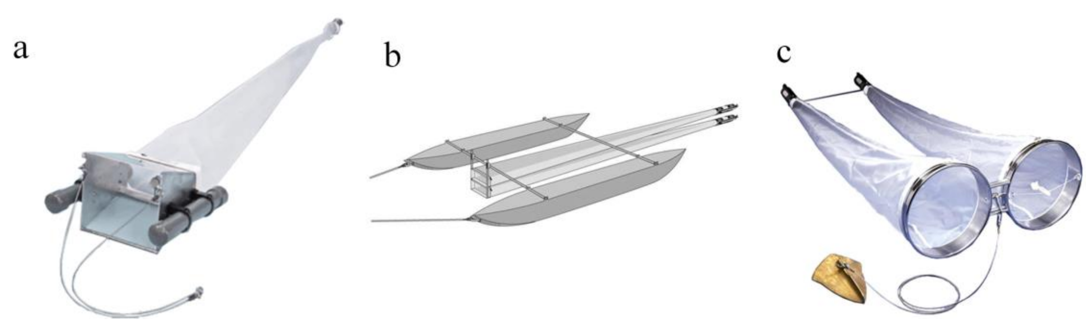

Figure 1. Diagram of trawling nets: (a) Manta trawl; (b) Neuston trawl, and (c) Bongo trawl.

The trawling net's mesh size determines the trapped particles' size and filtered water volume. Because of the risk of blockage for small meshes, trawling nets with a mesh size of approximately 330 μm are mostly used and accepted types (Lee, Shim, & Kwon, 2014; Eriksen, Mason, Wilson, Box, Zellers, Edwards, Farley, & Amato, 2013; Schönlau, Karlsson, Rotander, Nilsson, Engwall, Bavel, & Kärrman, 2020). Trawling nets allow for rapid filtration of large water volumes and collection of larger microplastic samples in a wider area compared with conventional pumping (Setälä, Magnusson, Lehtiniemi, & Norén, 2016). However, there are several defects for trawling nets, including 1) do not quantitatively sample microplastic particles < 300 μm (Covernton, Pearce, Gurney-Smith, Chastain, Ross, Dower, & Dudas, 2019; Barrows, Neumann, Berger, & Shaw, 2017); 2) require a towboat, which increases monitoring cost; 3) have limited ability to sample shallow or nearshore waters (Karlsson, Kärrman, Rotander, & Hassellöv, 2020); 4) the volume measurement of the filtered water may be inaccurate (Karlsson, Kärrman, Rotander, & Hassellöv, 2020); 5) have a slightly higher risk of contamination since the rinsing and transferring procedures from the collecting bag to the container after sampling usually take a relatively long time (Hidalgo-Ruz, Gutow, Thompson, & Thiel, 2012; Schönlau, Karlsson, Rotander, Nilsson, Engwall, Bavel, & Kärrman, 2020).

The pump has some advantages over trawling nets. Firstly, the pump does not need a towboat and is applicable for many platforms, such as ships, buoys, shore-based stations, etc. Secondly, the pump is more accurate in measuring the filtered water volume and is available for sampling in variable depths. Thirdly, the pump can collect microplastic of different sizes simultaneously with graded filter membranes (Schönlau, Karlsson, Rotander, Nilsson, Engwall, Bavel, & Kärrman, 2020). Nevertheless, the pump has shortcomings, such as the limited filtered water volume and the fixed-point sampling. The detailed comparisons for the trawling nets and the pump are listed in Table 1.

Table 1. Comparisons of the trawling nets and the traditional pump.

Sampling method	Time consumption	Contamination risk	Cost	Available platform	sampling volume
Trawling nets	long	large	boat necessary	ship only	large
Traditional Pump	short	small	boat unnecessary	ship、buoy、shore-based station	limited

Microplastic separation is usually accomplished by density flotation (or an improved method) (Imhof, Schmid, Niessne, Ivleva, & Laforsch, 2012; Nuelle, Dekiff, Remy, & Fries, 2014; Karlsson, Vethaak, Almroth, Ariese, Van Velzen, Hassellöv, & Leslie, 2017); filtration (Su, Xue, Li, Yang, Kolandhasamy, Li, & Shi, 2016); sieving (Baldwin, Corsi, & Mason, 2016), and pressurized fluid extraction (Fuller & Gautam, 2016). Since the sampling and separation steps are of paramount importance, standardized protocols should be established to avoid misidentification and underestimation of microplastics. The commercially available products are KC-Denmark's microplastic sampling pump and the German-SubCtech's sailing type system. These systems have a flexible mesh size range and can realize graded sampling of seawater microplastics. However, the seawater is filtered directly by the graded filter membranes, easily blocked by seawater organic materials. As a result, the pre-set water volume is probably affected or even incorrect. Therefore, there is an urgent need for a fast and easily implementable sampling system for seawater microplastics.

A novel *in situ* and graded sampling system for seawater microplastics was developed. Compared with the trawling nets, this system integrates the collection, separation, and enrichment procedures and features alternate water-air elution and three-layer graded collection (membrane mesh sizes 300, 100, and 50 μm). On the one hand, this system keeps the advantages of the traditional pump. On the other hand, this system is featured a T-type three-way filter assembly with a built-in filter element playing a buffer role, which is not possessed by the traditional pump. The filter element with a proper specific surface area protects the graded filter membranes from being blocked by the organic materials and guarantees the preset sampling water volume. A laboratory test was performed to assess the recovery of the sampling system. Then, an experimental verification was carried out at the Qingdao Coastal Test Station of the Institute of Oceanographic Instrumentation (Shandong Academy of Sciences) in January 2020 using the developed sampling system. The novel sampling system collected the seawater microplastics and accurately measured the filtered seawater volume.

Workflow of the sampling system

The workflow of the sampling system is illustrated in Figure 2: seawater microplastics are first enriched by the filter-element (316L quality stainless steel with mesh size 50 µm) in a T-type three-way filter assembly; then, microplastics in the filter-element are eluted alternately by air and seawater; finally, the graded collection of microplastics is achieved by the three-layer filter membrane of the graded collector (mesh sizes of 300, 100, and 50 µm from top to bottom). In Figure 2 the 3D model for the filter module of the sample system framed by the dashed box is also exhibited. The filter module consists of three parts: the T-type three-way filter assembly, the one-way valve and the graded collector, of which cross-section diagram is illustrated in Figure 3.

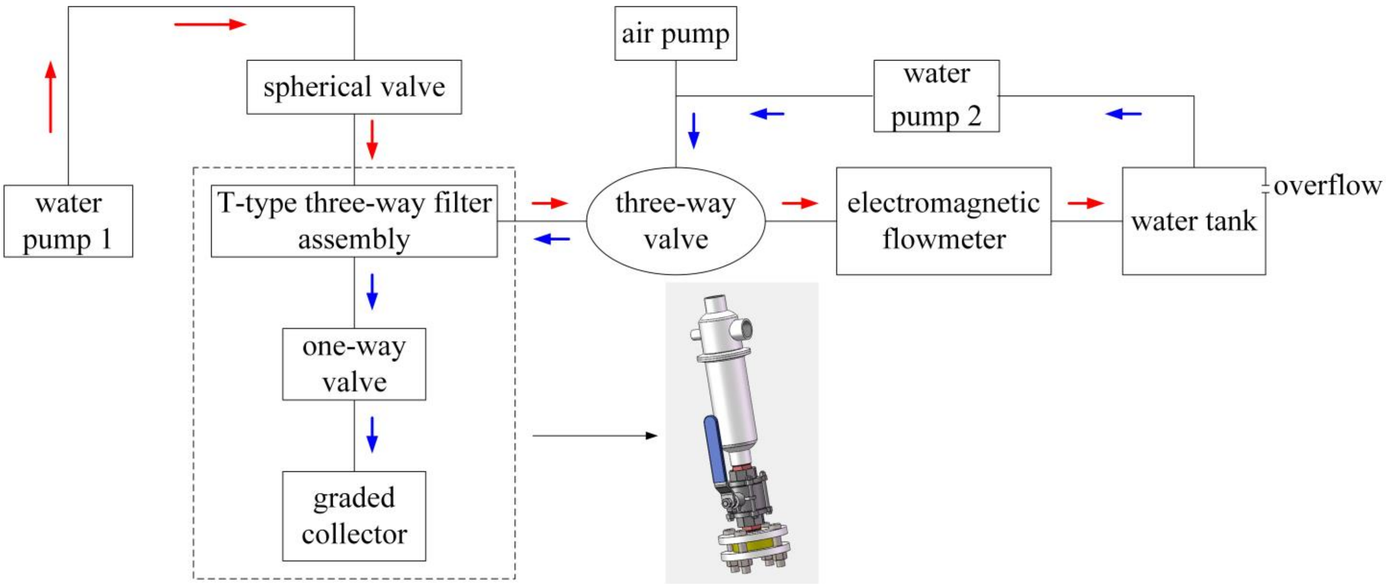

Figure 2. Workflow diagram of the sampling system.

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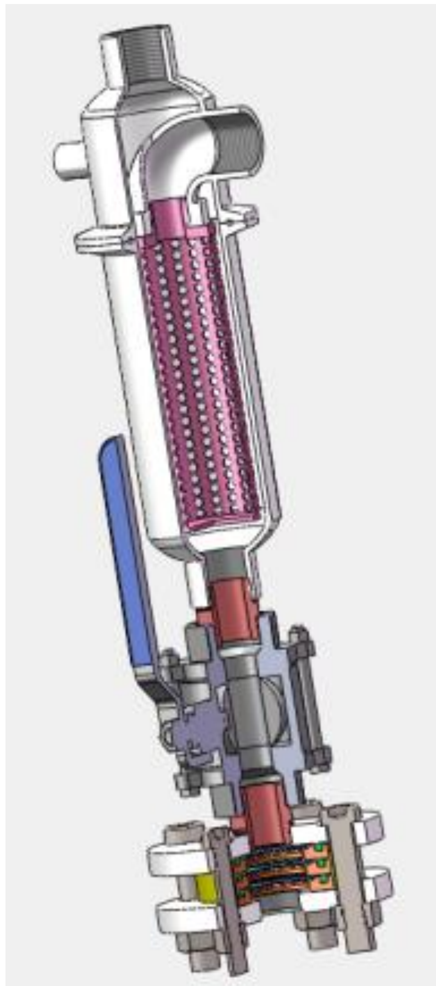

Figure 3. The cross-section diagram of the filter module.

Detailed operational procedures are as follows:

1) Enrichment. Open the spherical valve, close the one-way valve, open the three-way valve to connect the filter assembly to the electromagnetic flowmeter, and then open pump 1 (the bottom is installed with a filter with a mesh size of 5 mm to prevent large

pollutants from entering the collection device and blocking the pipeline). When seawater passes through the filter element of the filter assembly, microplastics are enriched; the filtered seawater passes through the three-way valve and the electromagnetic flowmeter and is then stored in a 60 l water tank with an overflow (shown by the red arrow line). The enrichment route could measure the filtered seawater volume accurately.

2) Elution. Close the spherical valve and open the one-way and three-way valve to connect the filter assembly with the air pump and water pump 2. Open pump two and flush the samples stored in the filter element of the filter assembly by using the filtered seawater (recycling seawater and avoiding the need for repeated pure-water flushing in the trawling method). Microplastics are eluted onto the graded collector's three layers of filter membranes. Then, open the air pump and flush the remaining seawater in the filter assembly onto the three-layer filter membrane. Elute 2–3 times by alternately using seawater and air to minimize the adhesion of the microplastics to the pipeline (shown by the blue arrow line). In practice, the elution route is usually completed in a certain period.

3) Graded collection. The eluted microplastics pass through the graded collector and are collected on the 300 μm, 100 μm, and 50 μm filter membranes according to their particle size.

Recovery assessment

As is known, the elution routine is set up to reduce the loss of microplastics for collection. The recovery performance of this system was evaluated by measuring the recovery rates with different elution times in the laboratory. A single 50 μm filter membrane of the graded collector was used for collection. At the beginning, the 50 μm filter membrane was weighted. Then, the microplastic samples with a known weight of W_1 were put into the system. After enrichment, elution, and collection, the 50 μm filter membrane was taken out and weighted. The weight of the microplastic samples collected on the filter membrane was W_2. The following equation estimates the recovery rate:

$$\text{Recovery rate} = \frac{W_2}{W_1} \times 100\ \%$$

The elution time is sequentially set to be 5, 10, 15, 20, 25, and 30 s, respectively. Under each elution time, the test was repeated three times. Figure 4 shows the recovery curve of the sampling system. It is seen from the figure that the recovery rate reaches more than 90 % in five seconds. Fluctuations of the recovery may be related to weighting errors or the uneven particle sizes of the inputted microplastic samples.

The overall recovery rate is above 80 %, which is better than the commonly accepted threshold of 70 % (Cashman, Ho, Boving, Russo, Robinson, & Burgess, 2020).

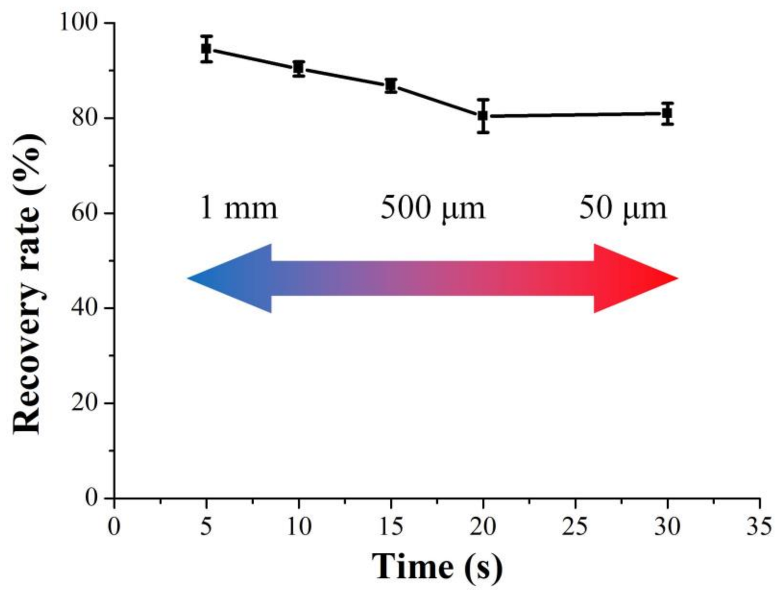

Figure 4. The recovery curve of the sampling system.

Materials and methods

A verification test was carried out by using the developed sampling system at the Qingdao Coastal Test Station (36° 3′ 28.7″ N, 120° 18′ 30.9″ E) of the Institute of Oceanographic Instrumentation (Shandong Academy of Sciences) in January 2020. As shown in Figure 5, Microplastic sampling was conducted at two adjacent positions. Exactly one-ton surface seawater was individually pumped at each position. In the experiment, an electromagnetic flowmeter was used to indicate the seawater's volume flowing through the sampling system. Figure 6 illustrates (a) the collected samples on the three-layer filter membranes and (b) their storage in watch glass to prevent contamination. At the same time, three blank filter membranes were put in the watch glasses as a set of blank controls, of which storage forms are similar to Figure 6(b). Then, the membranes containing microplastic samples and the blank controls were analyzed and identified through the same procedures.

Figure 5. The sampling location at Qingdao Coastal Test Station.

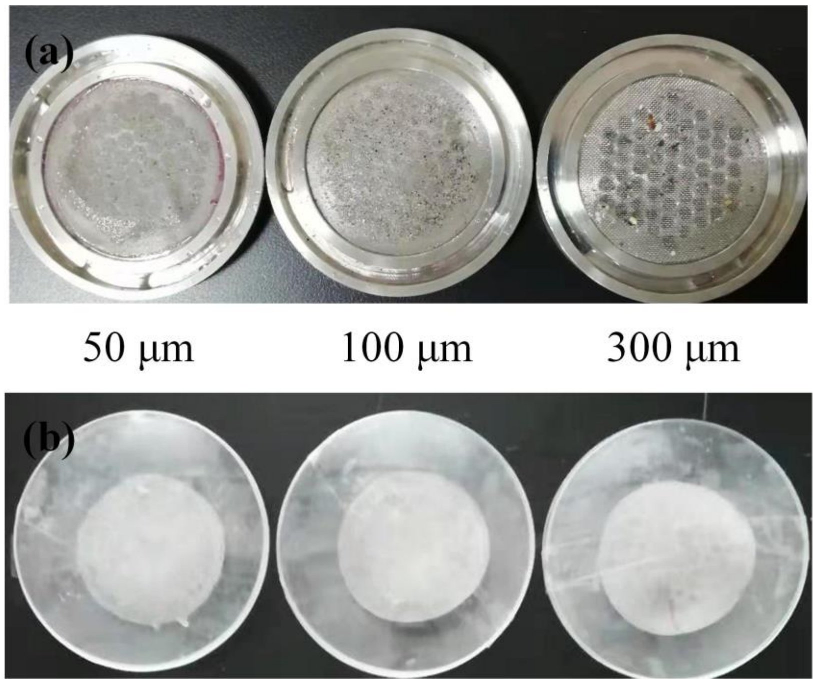

Figure 6. Illustrations of (a) the samples on the three-layer filter membranes and (b) their storage in a watch glass.

Instruments: Water pumps (model WQ (D) 4-10-0.55S, rated flow rate 4 m^3/h, rated lift 10 m, rated power 0.55 kW); a 6-caliber 316L spherical valve, one-way valve, three-way valve, and matching pipes; an air pump (model Michelin, voltage 12 V, pneumatic flow 25 l/min); an electromagnetic flowmeter; 316L stainless steel filter membranes (mesh sizes of 300, 100, and 50 µm); a Thermo Fisher DXR2 micro-Raman spectrometer; watch glass (70 mm), etc.

The digestion step was omitted during the process of microplastic identification for two reasons: one is that there are fewer algae in winter seawater, and the other is that the addition of chemical substances (such as H_2O_2) may change the morphological characteristics (size or color) of microplastics. (For example, 35 % hydrogen peroxide has already been confirmed to cause the discoloration and apparent size losses for several microplastics (Nuelle, Dekiff, Remy, & Fries, 2014). As a result, these microplastics could not be correctly identified. Molecular spectroscopy, such as infrared spectroscopy (IR) and Raman spectroscopy (Primpke, Lorenz, Rascher-Friesenhausen, & Gerdts, 2017; Cincinelli, Scopetani, Chelazzi, Lombardini, Martellini, Katsotiannis, Fossi, & Corsolini, 2017), is the most commonly used method for microplastic identification. Compared with IR spectroscopy, Raman techniques show better spatial resolution (down to 1 μm, while IR is 10–20 μm), wider spectral coverage, higher sensitivity to nonpolar functional groups, lower water interference, and narrower spectral bands (Araujo, Nolasco, Ribeiro, & Ribeiro-Claro, 2018). Therefore, in this context, micro-Raman spectroscopy (Thermo Fisher DXR2, laser wavelength 785 nm) was adopted to identify the microplastic samples (Zhao, Danley, Ward, Li, & Mincer, 2017; Zhang *et al*., 2017b). The acquisition exposure time was 20 seconds, the photobleaching time was one minute, and the spectral range was 50-3 500 cm^{-1}. The experimental parameter settings are listed in Table 2. Row-by-row scanning was first performed on the samples, and then the scanned spectra were compared with libraries of known spectra. If the matching degree was larger than 70 %, the particles were considered to be microplastics; their compositions were also determined (Mason, Welch, & Neratko, 2018).

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Table 2 Experimental parameter settings of Thermo Fisher DXR2.

Resolution ratio	Laser wavelength & energy	Acquisition exposure time	Photo bleaching time	Sample exposure	Spectral range
5 cm^{-1}	785 nm/10 mW	20 s	1 minute	2 times	50-3 500 cm^{-1}

Results

Microplastics were successfully collected by the system for these two sampling processes. The physicochemical properties of the microplastics, including the size, shape, color, and chemical composition, were further investigated. The blank control group was examined first, and no microplastics were found. This result means that the analysis procedures were not affected by potential sources of contamination in the environment. Microplastics were found on the 100 μm and 50 μm filter membranes, as shown in Figure 7 and Figure 8, respectively. On the 100 μm filter membrane (Figure 7), five types of microplastics: (a) polyester (PE); (b) polyethylene-vinyl acetate (PEVA); (c) and (d) polyethylene terephthalate (PET); (e) polyamide (PA), and (f) polystyrene (PS) were identified after matching their Raman spectra with the standard spectrum library. Their shapes can be divided into four categories: (a) foam; (b) and (d) fragment; (c) fiber; (e) and (f) granule. Their colors include translucent, white, blue, and yellowish. On the 50 μm filter membrane (Figure 8), four types of microplastics were identified: (a) PS, (b) PA, (c) poly(vinyl stearate) (PVS) and (d) PET. Although the microplastics on the two membranes may have the same composition, for example, PA, their shapes or colors are usually different. Information on type, color and shape for the above microplastics are summarized and listed in Table 3. These microplastics are similar to those reported at offshore water of Qingdao (Ding, Li, Sun, He, Jiang, Gao, & Zheng, 2018; Luo, Lin, Jia, Xu, & Li, 2019).

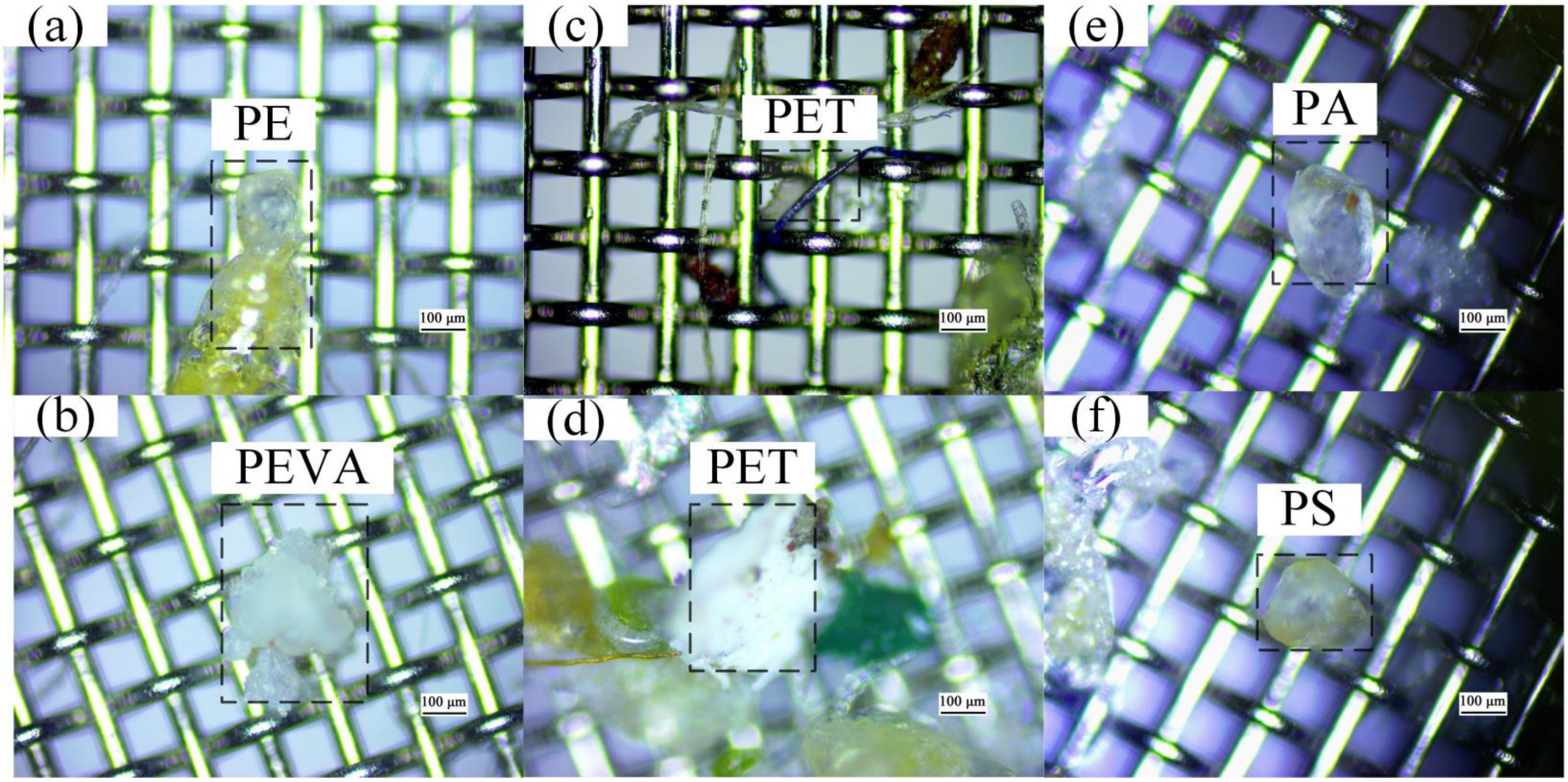

Figure 7. Microplastics on the 100 µm filter membrane.

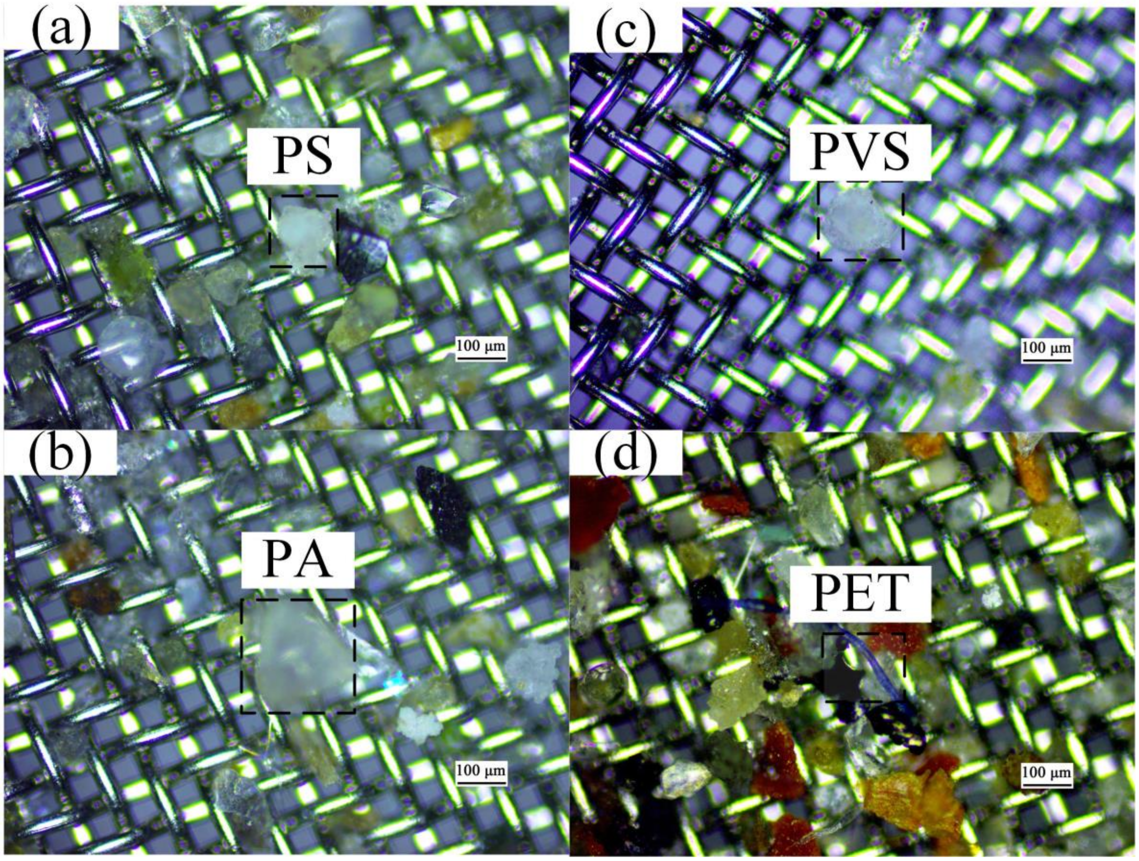

Figure 8. Microplastics on the 50 μm filter membrane.

Table 3. Microplastics information collected on the filter membranes.

Mesh size	MPs	Type	Color	Shape
100 μm	existence	PE、PEVA、PET、PA、PS	translucent、white、blue、yellowish	Foam、fragment、fiber、granule

50 µm	existence	PS、PA、PVS、PET	Blue、white、translucent	granule、fiber、foam

Figure 9 illustrates the characteristic Raman spectra and the corresponding standard spectra for these plastic polymers. However, no microplastics were found on the 300 µm filter membrane. The suggested reason is that microplastics with larger particle sizes (> 300 µm) are probably degraded into smaller particles by physical, chemical and microbial actions (Isobe, 2016). The above results are consistent with the conclusion in published studies that the abundance of microplastics decreases as their particle size increases (Tang, Liu, Zhou, He, Chen, Zhang, Hu, Huang, Luo, Ke, Chen, Xu, & Cai, 2018; Cai, He, Liu, Li, Tang, Wang, Huang, Wei, Lin, Chen, Hu, & Cen, 2018). It is harmful to the marine environment, because a number of experimental studies have demonstrated that the ingestion of microplastics with smaller particle size by aquatic species affects their growth, offspring quantity and behavior (Lo & Chan, 2018; Zhang, Wang, Song, Fang, & Wang, 2020). Since the sampling area is located near a bathing beach area, where human activities are frequent, tourism and textiles are guessed to be the main source of these microplastics (Andrady, 2011). Therefore, the microplastics may be prevented by several approaches, such as strengthen publicity and education, change consumption patterns, or develop environment-friendly materials.

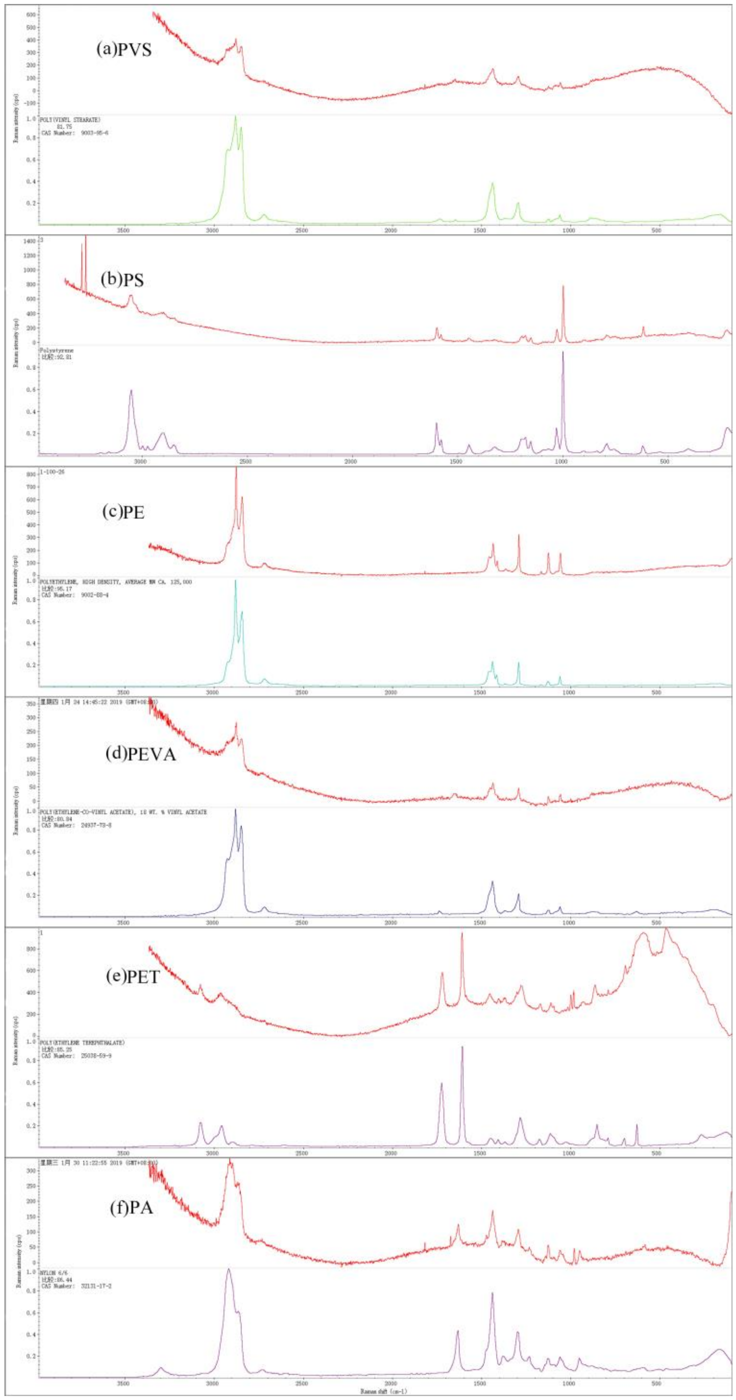
(a)PVS
(b)PS
(c)PE
(d)PEVA
(e)PET
(f)PA

Figure 9. Characteristic Raman spectra and the corresponding standard spectra for these plastic polymers: (a) PVS (b) PS, (c) PE, (d) PEVA, (e) PET and (f) PA.

Discussion

Microplastics with different types, colors and shapes were found on the 100 μm and 50 μm filter membranes. These results indicate the appropriateness of the sampling system for the *in situ* and graded collection of microplastics, especially those omitted by the trawling net method. In the proposed sampling system, the mesh size of the T-type three-way filter membrane was 50 μm. Therefore, microplastics with particle size larger than 50 μm were enriched to the T-type three-way filter surface. In theory, the system could work for the collection of microplastic greater than 300 μm. The reason why microplastic > 300 μm were not found is probably that larger microplastic do not exist (maybe degraded into smaller particles by physical, chemical and microbial actions) in the seawater during the sampling period.

Compared with trawl sampling, this system integrates the process of collection, separation and enrichment, which greatly reduce the operation time and avoid sample contamination. Another advantage of this system is that it features alternate air-water elution, seawater recycling and three-layer graded collection, which is not possible with the trawl.

Therefore, this sampling system provides the potential for the standardized sampling of microplastics in marine environmental monitoring. In theory, the measurable particle size of this sampling system ranges from 50 μm to 5000 μm. Notably, a more flexible mesh size range could be realized by replacing the filter membranes in the graded collector according to the demond, which lays a foundation for the collection of microplastics at a few microns or even nanoscale in the future.

Conclusions

In summary, a novel sampling system for the *in situ* and automatic graded collection of seawater microplastics was developed. This system integrates the collection, separation and enrichment procedures and features alternate air-water elution and three-layer graded collection (membrane mesh sizes of 300, 100 and 50 μm). Moreover, it is capable of measuring the filtered seawater volume accurately. An in-shore experiment was carried out to collect microplastic samples with the new system. These samples were analyzed and identified by micro-Raman spectroscopy. Main conclusions can be summarized as follows:

The advantages of the sampling system are ease of operation, reduced operation time, seawater recycling, and the formation of a closed space to avoid sample contamination. The sampling system is appropriate for the *in situ* and graded collection of microplastics, especially those omitted by trawling nets. This sampling system can be used as a supplement to the trawling net method and lays a foundation for the collection of microplastics at a few microns or even nanoscale in the future. This sampling system provides the potential for the standardized sampling of microplastics in marine environmental monitoring. The recovery rate of the sampling system reaches more than 90 % in five seconds, satisfying the requirements of rapid and efficient enrichment in conventional microplastics monitoring.

It should be pointed out that the experiment was initially designed to test the performance of the sampling system at one station. At present, valve control programming, core component optimization and other related procedures are being carried out to make the sampling system more portable and automatic. Moreover, multistation experiments are planned to be conducted by utilizing the optimized system to investigate the concentration, particle size distribution, type of polymer and distribution across the nearby sea of the microplastics. The expected research results can provide an effective technical means for the monitoring and early warning of microplastics pollution in Qingdao coastal area.

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