



Occurrence of microplastics in the seawater and atmosphere of the South China Sea: Pollution patterns and interrelationship



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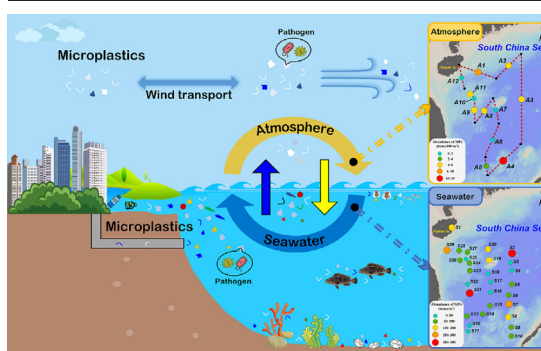
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HIGHLIGHTS

- Microplastics (MPs) are prevalent in seawater and atmosphere in the South China Sea
- PET fibers were dominant among the seawater and atmospheric MPs
- Seawater MP pattern was affected by anthropogenic activities and surface current
- Atmospheric MP pattern was affected by anthropogenic activities and wind conditions
- Seawater MPs had higher diversity and more complex sources than that in atmosphere

GRAPHICAL ABSTRACT



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ABSTRACT

Microplastic (MP) pollution is a serious global environmental problem, particularly in marine ecosystems. However, the pollution patterns of MPs in the ocean and atmosphere, particularly the sea-air interrelationship, remain unclear. Therefore, the abundance, distribution patterns, and sources of MPs in the seawater and atmosphere of the South China Sea (SCS) were comparatively investigated. The results showed that MPs were prevalent in the SCS with an average abundance of 103.4 ± 98.3 items/ m^3 in the seawater and 4.62 ± 3.60 items/ $100 m^3$ in the atmosphere. The spatial analysis indicated that the pollution patterns of seawater MPs were mainly determined by land-based discharge and sea surface currents, whereas atmospheric MPs were predominantly determined by air parcel trajectory and wind conditions. The highest MP abundance of 490 items/ m^3 in seawater was found at a station near Vietnam with current vortices. However, the highest MP abundance of 14.6 items/ $100 m^3$ in the atmosphere was found in air parcels with low-speed southerly winds from Malaysia. Similar MP compositions (e.g., polyethylene terephthalate, polystyrene, and polyethylene) were observed in the two environmental compartments. Furthermore, similar MP characteristics (e.g., shape, color, and size) in the seawater and atmosphere of the same region suggested a close relationship between the MPs in the two compartments. For this purpose, cluster analysis and calculation of the MP diversity integrated index were performed. The results showed an obvious dispersion between the two compartment clusters and a higher diversity integrated index of MPs in seawater than in the atmosphere, thus implying higher compositional diversity and more complex sources of MPs in seawater relative to the atmosphere. These findings deepen our understanding of MP fate and patterns in the semi-enclosed marginal sea environment and highlight the potential interrelationship of MPs in the air-sea system.

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1. Introduction

The pollution caused by microplastics (MPs), plastic debris smaller than 5 mm, has become a serious global environmental concern owing to their ubiquity and potential threats to ecosystems (Rochman, 2018). Owing to the cost-effectiveness and convenience of plastics, their production and use have increased sharply in recent decades (Reisser et al., 2015). The global plastic output was estimated as 367 million metric tons (Mt) in 2021 (Plastics Europe, 2021). However, approximately 275 million Mt. of plastic waste is generated annually due to poor management and the low recycling capacity of plastic waste (Jambeck Jenna et al., 2015). Through ultraviolet radiation, weathering, and microbial degradation, plastic debris in the natural environment can easily fracture into small particles such as MPs and nanoplastics (Arthur et al., 2009; Thompson Richard et al., 2004). Most MPs are secondary MPs generated by the fragmentation of larger plastic debris (Gardon et al., 2021). In contrast, primary MPs are released directly into the environment as micro-sized particles, such as synthetic fibers, cosmetic microbeads, pre-production pellets, and tire particles (Cui et al., 2022).

Owing to their buoyancy and small-size, MPs are readily ingested or inhaled by filter-feeding zooplankton (Wang et al., 2020c), bivalves (Liu et al., 2021a), fish (Wang et al., 2020d), and even humans (Prata, 2018), probably causing physical (e.g., digestive system blockage, reduction in the feeding rate and energy reserves, and gastrointestinal abrasions) and ecotoxicological damage (e.g., oxidative stress, inflammatory responses, neurotoxicity, reproductive toxicity, cellular damage) and cardiopulmonary disease (Bringer et al., 2020; Liu et al., 2021a; Prata, 2018; Wang et al., 2020d). Extensive studies have also revealed the potential roles of MPs as vectors of organic pollutants (e.g., PAHs and PCBs) and heavy metals, thus posing more complex ecotoxicological risks (Ding et al., 2022; Torres et al., 2021). Moreover, MPs can be colonized by different microbial communities, thus forming “plasticsphere,” a new habitat for microbes, especially pathogens, and possibly exacerbating the spread of epidemics (Amaral-Zettler et al., 2020; Arias-Andres et al., 2018; Viršek et al., 2017; Yang et al., 2018). Therefore, MP pollution in the natural environment is a persistent concern.

Transported by various channels, such as wastewater effluents, runoff, rivers, and improper disposal, an increasing number of MPs inevitably enter the ocean from terrestrial and coastal ecosystems (Kooi et al., 2016). In addition to land-based sources, MPs can be transported into the ocean through atmospheric deposition (Liu et al., 2019b). Once in the marine environment, MPs generally float in seawater and water columns or sink to the seafloor, depending on their density and biological interactions (Porter et al., 2018). Buoyant MPs can be influenced by water currents, wind, tides, and anthropogenic activities (e.g., fishing and shipping) to undergo horizontal transportation (Iwasaki et al., 2017; Rocha-Santos and Duarte, 2015). They can also be colonized by organisms (Sun et al., 2018), incorporated into marine snow (Porter et al., 2018), or ingested by marine organisms (Wang et al., 2020c), causing vertical migration. Notably, MPs can also be discharged into the atmosphere through sea wave emissions (Ferrero et al., 2022). As the largest source of aerosols in the atmosphere, sea wave emissions can discharge large amounts of MPs from seawater into the atmosphere under the action of strong winds (Allen et al., 2020; Spada et al., 2013). In the past, many studies focused on MP pollution in oceans such as the Pacific Ocean (Egger et al., 2020; Liu et al., 2021b), the Arctic Ocean (Kanhai et al., 2020), the Indian Ocean (Li et al., 2021b; Patti et al., 2020), the German North Sea (Roscher et al., 2021), the Atlantic Ocean (Silvestrova and Stepanova, 2021), Arctic and Antarctic snow (Aves et al., 2022; Bergmann et al., 2019), and in the marine boundary layer atmosphere such as over the Pacific Ocean (Liu et al., 2019b), the Indian Ocean (Wang et al., 2020b), Baltic Sea (Ferrero et al., 2022), and Yellow Sea (Liu et al., 2020). These studies indicated the extensive occurrence and potential long-range transport of MPs. However, knowledge of the MP pollution status and distribution patterns in the ocean and its marine boundary layer atmosphere, especially the sea-air exchange of MPs, is still limited.

The South China Sea (SCS) is the largest marginal sea in China. It is located in the western Pacific Ocean and contains rich resources for marine oil and gas, coastal and island tourism, marine energy, port shipping, and biodiversity (Barber and Pratt, 1998; Xu et al., 2019). As one of the most important tropical ecosystems in the world containing islands, coral reefs, mangroves, and seagrass beds, the SCS provides a suitable habitat for marine species in various niches (Huang et al., 2015; Sun et al., 2017). However, the SCS is under nonnegligible anthropogenic pressure from circumjacent developing countries (e.g., the Philippines, Malaysia, Vietnam, and China) with large populations, numerous industries, agriculture, aquaculture, and shipping (Cai et al., 2018). Owing to the lack of effective recovery and disposal policies and measures, improper coastal landfilling and dumping are significant sources of xenobiotic pollutants in the SCS (Huang et al., 2019; Jambeck Jenna et al., 2015). Increasing evidence has shown that the marine ecosystems of the SCS are heavily contaminated by emerging pollutants such as organophosphate esters (Ding et al., 2020), organochlorine pesticides (Ding et al., 2019b; Kang et al., 2022), perfluoroalkyl substances (Kwok et al., 2015; Pan et al., 2018), antibiotics (Zhang et al., 2019; Zhang et al., 2018), and MPs (Huang et al., 2019; Nie et al., 2019; Tan et al., 2020). However, the distribution pattern and potential sources of MPs in the SCS remain unclear because of the low coverage of study areas and inconsistent methods.

Thus, samples of surface seawater and marine boundary layer atmosphere were collected simultaneously in the SCS. The specific objectives of this study were to 1) investigate the abundance, characteristics, distribution patterns, and possible sources of marine and airborne MPs and 2) illustrate the interrelationship of MPs between the two compartments. This study provides baseline data on MPs in the SCS to understand the health status of semi-enclosed marginal sea ecosystems and to distinguish MP sources and potential interrelationships between airborne and marine MPs.

2. Materials and methods

2.1. Study area and sample collection

The SCS is a marginal sea that is largely surrounded by land such as China, Malaysia, the Philippines, and Vietnam. It has been studied in numerous islets, atolls, and reefs. The SCS covers an area of approximately 3.3 million km² and ranges in depth from the shallowest coastal fringe to 5377 m in the Manila Trench. It occupies the northern tropics and experiences a monsoonal climate influenced by the southwest monsoon in summer and the northeast monsoon in winter (Morton and Blackmore, 2001). Sampling was conducted during an oceanographic research cruise in the SCS between May and June 2021. Surface seawater and atmospheric samples were collected between 8–19° N and 110–116° E. Information on the surface seawater sampling stations and the atmospheric sampling track is shown in Fig. S1 and Table S1.

Surface seawater MPs were collected at a depth of 0.2 m using a stainless-steel suction filter pump (Huang et al., 2019; Nie et al., 2019). A total of 29 seawater samples were collected in this study, of which S1 was collected from a harbor on Hainan Island. The pumping system was submerged to a depth of approximately 0.1–0.2 m. The pumped seawater was allowed to flow through a stainless-steel pipe and was concentrated with a 64- μ m plankton net. After collecting 200 L of seawater, the sieve was rinsed at least thrice with Milli-Q water. The concentrated sample in the collection tube at the end of the sieve was immediately filtered through a nylon filter (Millipore, 20 μ m, 47 mm) using a glass filtration device. The filters were then carefully placed horizontally in a glass Petri dish wrapped with aluminum foil and stored at 4 °C prior to further laboratory analyses.

Suspended atmospheric MPs were collected along the cruise track using an atmospheric total suspended particulate sampler (Jinshida, 100 \pm 0.1 L/min) (Wang et al., 2020b; Wang et al., 2021). The sampler was placed horizontally on the upper deck at a height of 1.5 m to prevent pollution from the ship. Twelve atmospheric samples were collected, all of which were of low-altitude suspended atmospheric MP origin. Each atmospheric sample was collected for 12–24 h (67.1–129 m³ of air) through a 1.6- μ m pore size

glass microfiber filter (Whatman, GF/A) depending on the meteorological conditions. After collection, the filters were placed horizontally in a glass petri dish, wrapped with aluminum foil, and stored at 4 °C before being transferred to the laboratory. The sampling date, volume of filtered air, wind speed, wind direction, humidity, temperature, and air pressure during the sampling period were recorded synchronously because the distribution pattern of atmospheric MPs can be easily affected by meteorological factors.

2.2. Isolation, observation, and identification of MPs

For the seawater samples, MPs were extracted by Fenton oxidation (Prata et al., 2019). First, the particles concentrated on the nylon filters were digested for 48 h using Fenton's reagent (30% H₂O₂ with a ferrous iron catalyst) at 65 °C. Subsequently, the digestion solution (100 mL/sample) was filtered through a 1.6-μm glass fiber filter. Seawater and atmospheric samples on glass fiber filters were visually classified using a stereo microscope at a 20–160 × magnification (Nikon SMZ1270, Japan) and identified according to preset criteria (Hidalgo-Ruz et al., 2012) (see Text S1, *Supplementary Information*). Images of the identified MP particles were obtained using an IMG SC2000C digital camera, and their size, color, and shape were recorded. After visual identification, all the suspected MP particles (≥ 20 μm) were identified by PerkinElmer Spectrum Spotlight 200i micro-Fourier transform infrared spectroscopy (μ-FTIR) (PerkinElmer, USA). The spectrum ranged from 4000 cm⁻¹ to 600 cm⁻¹ with a spectral resolution of 8 cm⁻¹. The obtained spectra were compared with the standard spectra in the software database, and a similarity of > 65% was accepted as a polymer match. The surface structures of MPs from seawater (before digestion; 1.48%, nine particles) and the atmosphere (8.34%, five particles) were further analyzed using scanning electron microscopy (SEM, TESCAN VEGA3, Czech Republic). Notably, the MP samples were fixed on carbon tabs and sputtered with a 3 nm layer of gold under vacuum to enhance conductivity.

2.3. Backward trajectory analysis

The Hybrid Single Particle Lagrangian Integrated Trajectory backward trajectory model was used to analyze the potential source routes of atmospheric MPs (Allen et al., 2019). The data from the Global Data Assimilation System covered the atmospheric sampling period, and the duration of each trajectory was 72 h. The starting height of the trajectory was set to a sampling height of 10 m. After model simulation, the generated backward trajectory maps of the 12 sampling transects were imported into ArcGIS.

2.4. MP diversity integrated index analysis

The MP diversity integrated index (MDII), which is constructed based on Simpson's diversity, can be used to analyze the associations and differences between MPs in different environmental units or compartments, reflecting the uniformity and richness of MP characteristics as well as the number of MP pollution sources (Zhou et al., 2022). Herein, the MDII calculated based on four Simpson diversity indices (SDI), including size (SDI_S), shape (SDI_{SH}), color (SDI_C), and polymer type (SDI_{PT}), was used to reflect the diversity of MP composition and the number of MP pollution sources (Li et al., 2021a). The formula for calculating MDII is as follows:

$$P_i = \frac{N_i}{N} \quad (1)$$

$$SDI = 1 - \sum_{i=1}^N P_i^2 \quad (2)$$

$$MDII = \sqrt[4]{(SDI_S \times SDI_{SH} \times SDI_C \times SDI_{PT})} \quad (3)$$

where N represents the number of categories and i represents the index of categories from 1 to N . MDII ranges from 0 to 1; the closer the value is to 1, the higher the diversity of MP composition and the number of MP pollution sources.

2.5. Quality assurance and quality control

All containers, including glass Petri dishes, glass conical bottles, blue-cap glass bottles, and sample storage boxes, were washed at least three times with Milli-Q water before use and wrapped with aluminum foil to avoid atmospheric MP pollution. A stereomicroscope for visual classification was placed on a glass cover, and both hands entered from the operating hole during microscopy. Nitrile gloves and cotton lab coats were worn during the sampling and experimental processes. Procedural blanks were collected and treated synchronously with the field samples to correct for and evaluate background pollution. Procedural blanks for seawater samples were obtained by replacing seawater with 500 mL of Milli-Q water, and procedural blanks for atmospheric samples were prepared by placing a clean filter in the atmospheric sampler for 1 h while the power was turned off. Overall, only one piece of polyethylene terephthalate (PET) fiber (transparent, 1330.88 μm) was found in the seawater procedural blank, and no MP contamination was observed in the atmospheric procedural blank. The abundance of MPs in seawater was corrected for procedural blanks.

2.6. Statistical analysis

The abundances of MPs in seawater and the atmosphere were expressed in items/m³ and items/100 m³, respectively. The data were analyzed using Microsoft Excel 2016 and SPSS 25.0. Graphs were created using Ocean Data View 5.1.5, ArcGIS 10.2, R 3.5.1, and Excel 2016. The relationships between seawater and atmospheric MPs and between atmospheric MP abundances and meteorological factors were explored using Principal Component Analysis (PCA) and checked using Spearman's correlation analysis. The differences in the MDII of seawater and atmospheric MPs, MP size, and average abundance were determined using the Kruskal-Wallis test. In all tests, statistical significance was set at $P < 0.05$.

3. Results and discussion

3.1. Occurrence and characteristics of MPs in seawater

MPs were detected in all seawater stations with an average abundance of 103.4 ± 98.3 items/m³ (Fig. 1a, Table S1), showing that the seawater in the SCS was extensively contaminated by MPs. As shown in Table S3, the MP abundances in worldwide surface seawater differed by several orders of magnitude, mainly because of differences in the sampling area, mesh size, volume of filtered seawater, environmental conditions (e.g., wind and current), sampling date, and laboratory analytical methods (Constant et al., 2018; Liu et al., 2019c; Shim et al., 2018; Sun et al., 2021; Zheng et al., 2021). Therefore, the MP abundances in the present study were compared with those in studies using similar sampling methods. The results showed that the MP abundances (20–490 items/m³) in the seawater in this study were lower than those previously reported (1400–15900 items/m³) in the SCS (Cai et al., 2018; Ding et al., 2019a; Huang et al., 2019; Nie et al., 2019), but comparable to those (70.8–279 items/m³) in the northeastern Pacific Ocean and the North Atlantic Ocean (Courtene-Jones et al., 2017; Desforges et al., 2014).

More than twenty different polymer types were identified using μ-FTIR spectroscopy (Fig. 1a). Generally, PET, polystyrene (PS), polyethylene (PE), poly (propylene-ethylene) (PP-PE), poly methyl methacrylate (PMMA), and polyethylene glycol (PEG) were the major MPs in seawater. The FTIR spectra and images of the predominant polymer types of the MPs are shown in Fig. 2. Among the seawater samples, PET accounted for the highest proportion (34.0%), followed by PS, PE, PP-PE, PMMA, and PEG, which accounted for 21.2%, 7.22%, 5.42%, 5.42%, and 5.42%, respectively. Plastic products of PET, PS, PE, and PP-PE are widely applied in industries and households, such as textiles, containers, water bottles, packaging bags, electronics, toys, drinking straws, agricultural films, and automotive components (Auta et al., 2018; Inuwa et al., 2014; Zhu et al., 2018). These MP types were also frequently identified in the SCS by previous studies (Cai et al., 2018; Ding et al., 2019a; Huang et al., 2019). Notably, PMMA and PEG, which

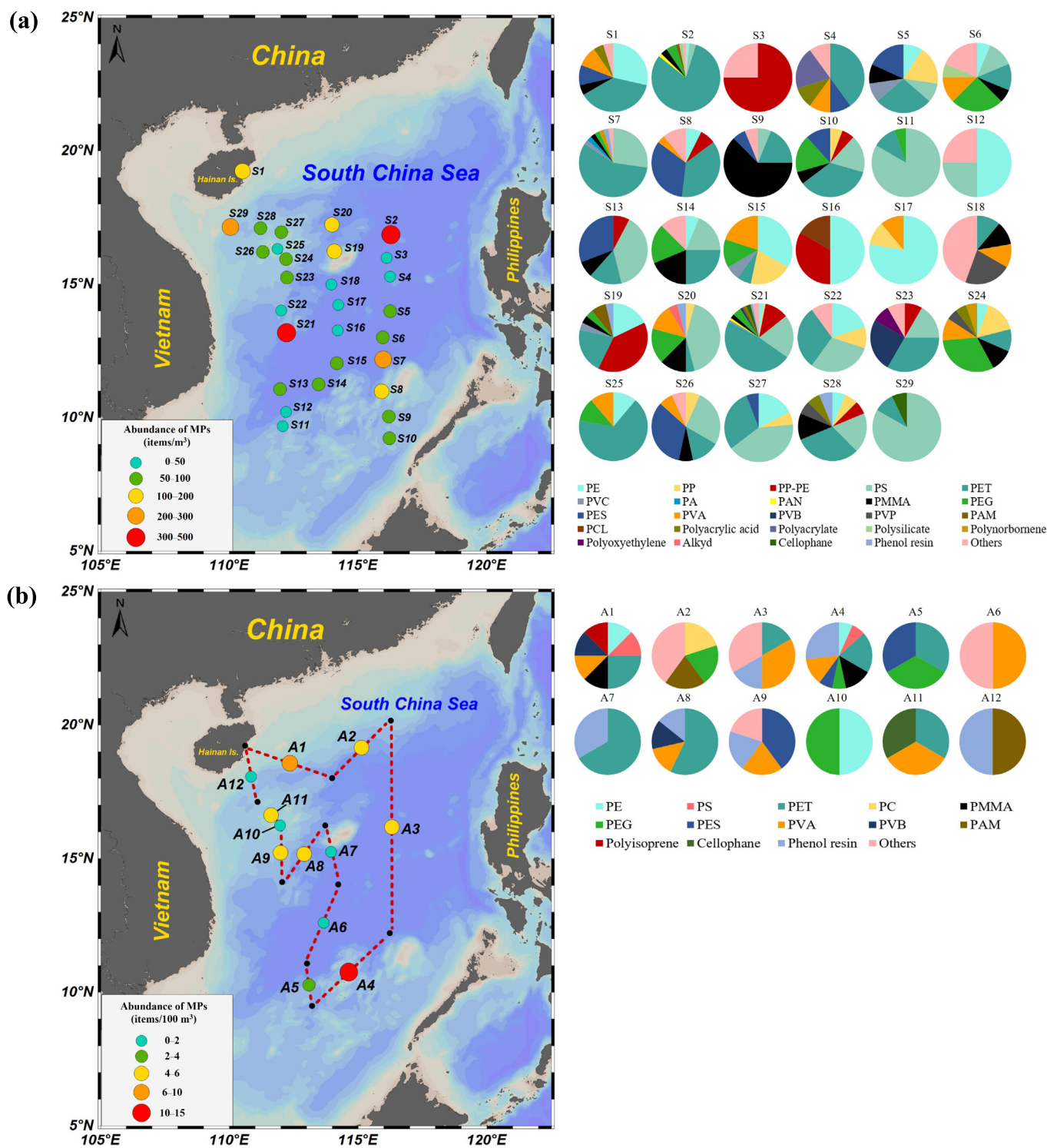


Fig. 1. The abundances and composition of MPs in seawater (a) and atmosphere (b) in the SCS. PE: polyethylene; PP: polypropylene; PP-PE: poly(ethylene-propylene); PS: polystyrene; PET: polyethylene terephthalate; PC: polycarbonates; PVC: polyvinyl chloride; PA: polyamide; PAN: polyacrylonitrile; PMMA: polymethyl methacrylate; PEG: polyethylene glycol; PES: polyester; PVA: polyvinyl alcohol; PVB: polyvinyl butyral; PVP: polyvinyl pyrrolidone; PAM: polyacrylamide; PCL: polycaprolactone.

are used in buildings, advertisements, decoration applications, and personal care products (Brahney et al., 2020; Wilkes and Aristilde, 2017), were identified in the present study but were rarely observed in the surface seawater of the SCS. These results suggest that the number of MP types has increased in recent years owing to the rapid growth in plastic use and poor management of plastic waste.

Four MP shapes including fibers, granules, fragments, and films, were detected in the seawater samples (Fig. 3a, S2a). Fibers were predominant (64.5%), followed by granules (27.9%), films (4.27%), and fragments (3.28%). Larger amounts of fibers may be derived from human textile washing and fishery activities (Xue et al., 2020). An experiment demonstrated that a single garment can produce > 1900 plastic fibers per wash

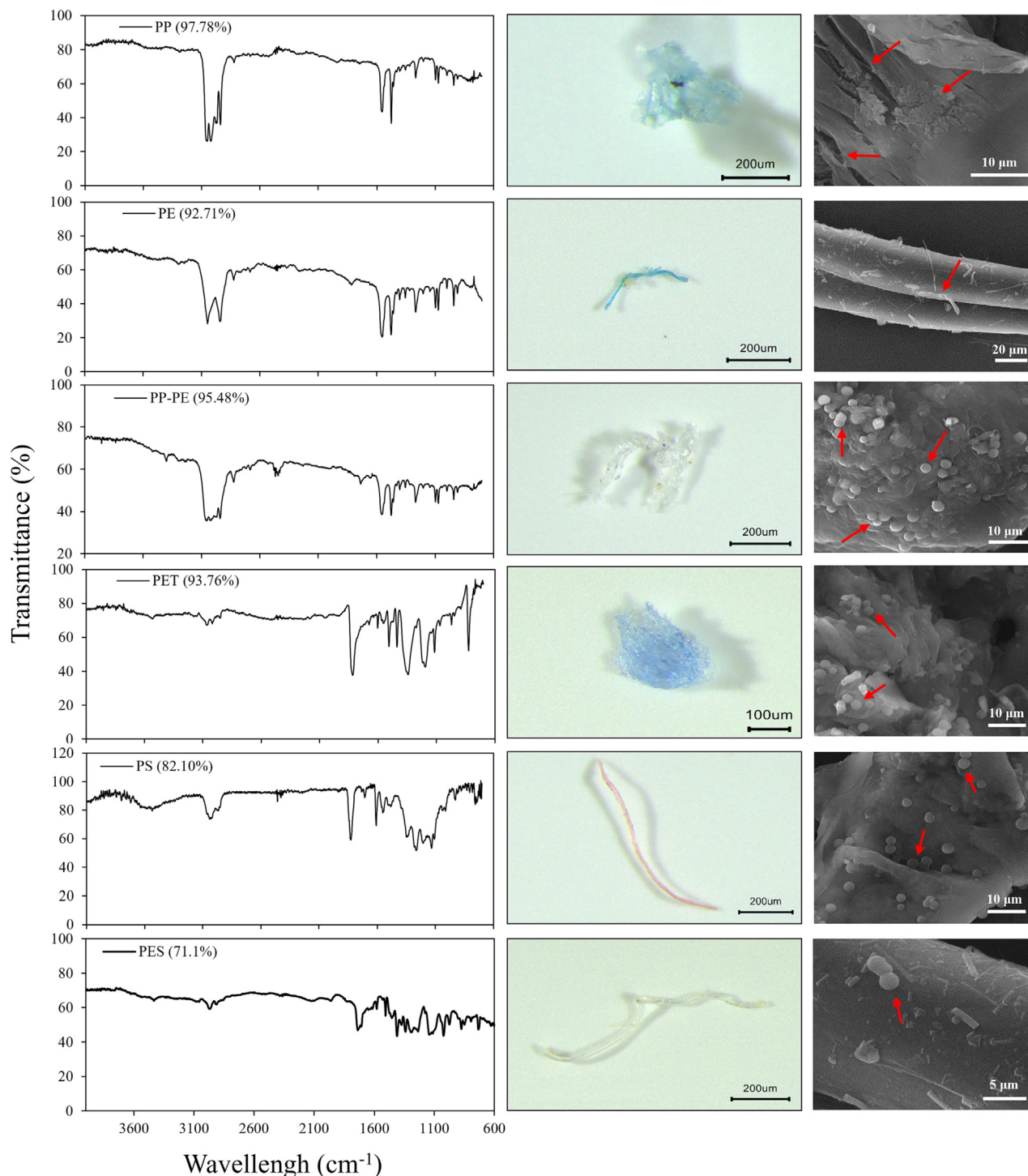


Fig. 2. Infrared spectra, and match degrees, microscope images and surface morphology of the representative MPs in seawater (PP, PE, and PP-PE) and atmosphere (PET, PS, and PES). The arrows represent cracks, organic matters, and the suspected microbes.

using domestic washing machines (Browne et al., 2011), and these plastic fibers are emitted into the ocean through coastal municipal wastewater (Murphy et al., 2016). Additionally, fishing gear such as ropes, nets, and pots, as well as the abrasion of gear through frequent fishing activities, may release MP fibers into the surrounding sea area (Huang et al., 2019; Nie et al., 2019).

Seven colors were observed for the MPs in seawater: red, black, yellow, green, blue, transparent, and white (Fig. 3c, S2b). Among them, transparent, blue, and green were the three dominant colors, accounting for 83.4% of all color classes. The transparent and blue colors are the most

common colors detected in all surveys of marine MPs (Cai et al., 2018; Jiang et al., 2020; Patti et al., 2020; Silvestrova and Stepanova, 2021; Wang et al., 2020a). The size range of the seawater MPs was 34.2–4996 μm with an average size of 669 μm . The sizes of < 50, 50–100, 500–1000, and 1000–5000 μm accounted for 1.97%, 4.93%, 26.6%, and 18.6%, respectively, and the 100–500 μm size fraction (47.9%) was the most prevalent among all the size classes (Fig. 3e), similar to those in the western Pacific Ocean (Zhang et al., 2020). Additionally, no MPs smaller than 30 μm were identified in this study, mainly because of the large mesh size of 64 μm used for pre-concentration during sampling. It is

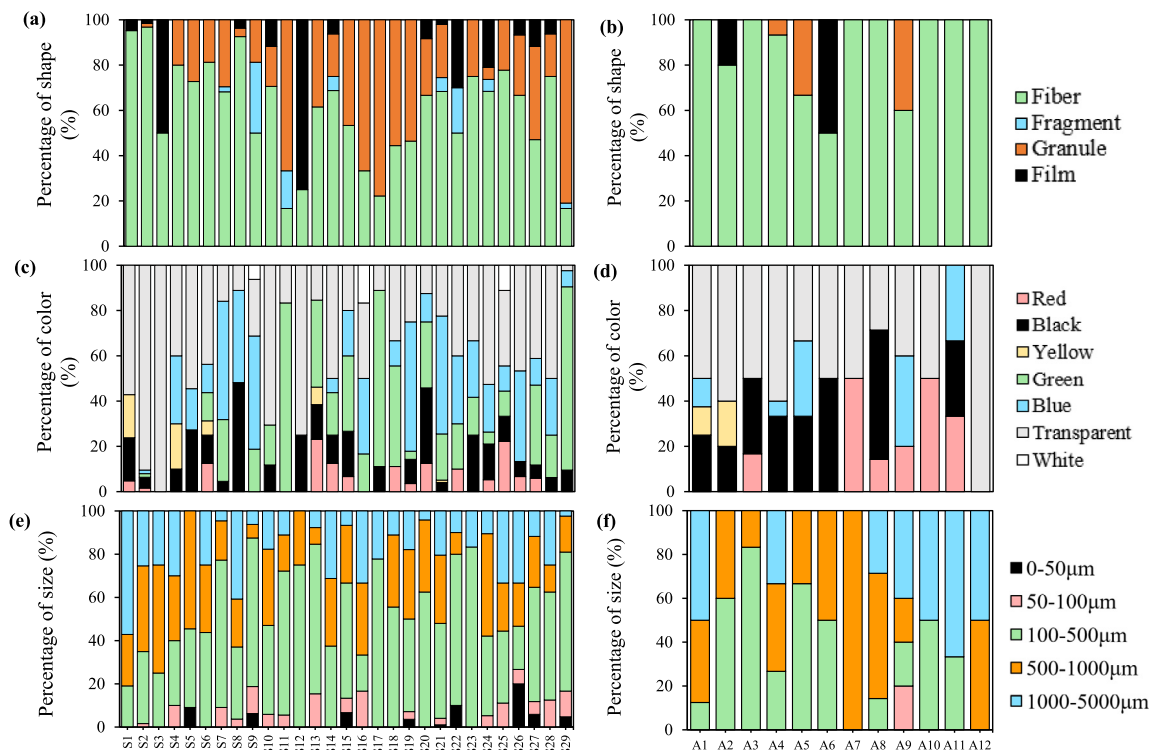


Fig. 3. Shape, color and size distribution of MPs in seawater (a, c, and e) and atmosphere (b, d, and f).

worth noting that MPs with sizes of 34.2–330 µm accounted for 36.5% of the total seawater MPs. However, these MP fractions are easily ingested by bivalves and fish owing to their similar size to algae (Lindeque et al., 2020) and cannot be collected using plankton nets with mesh sizes of 330 µm or 333 µm for sampling (Cai et al., 2018). These results imply that many previous studies have underestimated the pollution levels of marine MPs and the corresponding ecological risks, which should be solved by establishing standards for MP monitoring methods.

Several MP fibers, fragments, and films were randomly selected and analyzed by SEM. All the selected MPs exhibited obvious morphological features resulting from aging and mechanical weathering in the marine environment (Fig. 2). For example, cracks and fractures were observed on MP films, and organic matter attachment was observed on MP fibers. In addition, large numbers of spherical suspected microbial cells were found on the MP fragments (Fig. 2). The suspected microbial attachment could be explained by the stronger persistence of MPs (e.g., PET and PE) than that of natural buoyant matter (e.g., branches and feathers) (Chamas et al., 2020), while the hydrophobic surfaces of MPs provide favorable habitats for microbes (Zhang et al., 2022). Additionally, MPs and adsorbed nutrients can provide carbon and energy sources for microbial growth and metabolism (Paço et al., 2017). MPs may be carriers and reservoirs of human pathogens, including antibiotic resistant bacteria (ARB) and antibiotic resistance genes (ARGs) (Liu et al., 2021c; Pham et al., 2021; Yang et al., 2018), posing a nonnegligible threat to the health of marine ecosystems and humans.

3.2. Occurrence and characteristics of MPs in the atmosphere

MPs were detected in all 12 atmospheric samples. Atmospheric MPs were widely distributed in the cruise transects, with their abundances ranging from 1.55 items/100 m³ to 14.6 items/100 m³ (Fig. 1b, Table S2). As shown in Table S4, the average abundance of atmospheric MPs (4.62 ± 3.60 items/100 m³) in the present study was lower than those in the western Pacific Ocean (6 ± 16 items/100 m³) (Liu et al., 2019b) and the Yellow Sea (19 ± 13 items/100 m³) (Liu et al., 2020), but higher than those in the east Indian Ocean (0.4 ± 0.6 items/100 m³) (Wang et al., 2020b) and the

SCS (0.39 ± 0.43–3 ± 3 items/100 m³) (Liu et al., 2020; Wang et al., 2020b; Wang et al., 2021). These different results may be due to differences in weather conditions, sampling dates, and areas. Compared with marine environments, urban areas, such as Paris and Shanghai, have higher suspended atmospheric MP abundances of 30–150 items/100 m³ and 142 ± 142 items/100 m³, respectively (Dris et al., 2017; Liu et al., 2019a). These results confirm that extensive anthropogenic activities have aggravated atmospheric MP pollution. However, a study that collected both suspended and deposited MPs reported a relatively high abundance (2400 ± 900 items/100 m³) of atmospheric MPs in the open Baltic Sea (Ferrero et al., 2022). MP deposition (dry and wet deposition) is another important pathway of the global MP cycle and usually drives high MP pollution in urban environments, such as Guangzhou (114 ± 40 items/m² d) (Huang et al., 2021), Dongguan (175–313 items/m² d) (Cai et al., 2017), and Hamburg (136.5–512.0 items/m² d) (Klein and Fischer, 2019).

Sixty particles collected from the atmosphere were identified as MPs. In contrast to seawater samples, only 14 polymer types were detected in the atmospheric samples, fewer than that in seawater MPs (25 types) (Fig. 1b). The major polymer types in atmospheric samples were PET (21.7%), polyvinyl alcohol (PVA) (15.0%), phenol resin (15.0%), PEG (6.67%), and polyester (PES) (6.67%). These results are consistent with those of previous studies in the eastern Indian Ocean, Pearl River Estuary, western Pacific Ocean, and the SCS (Liu et al., 2019b; Wang et al., 2020b; Wang et al., 2021). The prevalence of PET and PES in the atmosphere is mainly due to their extensive use in the textile industry, e.g., in human clothing, curtains, furniture covers, and bed textiles (Dris et al., 2016; Hidalgo-Ruz et al., 2012). Phenol resin is a high-density polymer (1.17–2.00 g/cm³) and may be entrained to the atmosphere by severe turbulence, updrafts, and vertical mixing (Kedzierski et al., 2018; Wang et al., 2021).

Atmospheric MPs were identified in three shapes: fibers, granules, and films (Fig. 3b, S2a). Most atmospheric MPs were fibers (90.0%), with small amounts of granules (6.67%) and films (3.33%). Similarly, a high proportion of fibers (59.1–88.9%) was found in the suspended atmospheric MPs over the Pearl River Estuary, the east Indian Ocean, urban Shanghai (Liu et al., 2019a; Liu et al., 2019b; Wang et al., 2020b; Wang et al.,

2021), and the MP deposition in Guangzhou and Dongguan (Cai et al., 2017; Huang et al., 2021). This is likely due to the fact that fibers are easily separated from clothing and ornaments, and are generally lighter and easier to enter the atmosphere than other shapes (Ding et al., 2021). Atmospheric MPs consisted of five colors, with the most prevalent being transparent (48.3%) and black (28.3%) (Fig. 3d, S2b). It is noteworthy that all granules detected in our study were blue PES, suggesting similar sources. The size range of atmospheric MPs was 90.4–2684 μm with an average size of 763 μm (Fig. 3f). The average size of atmospheric MPs in the present study was larger than those in the northern SCS (466.04 \pm 409.47 μm) (Ding et al., 2021; Wang et al., 2021), east Indian Ocean (643.13 \pm 319.40 μm) (Wang et al., 2020b), and west Pacific Ocean (349.76 \pm 395.83 μm) (Liu et al., 2019b), but smaller than those in the Pearl River Estuary (917.38 \pm 563.60 μm) and SCS (953.00 \pm 730.10 μm) (Wang et al., 2020b). The predominant sizes of MPs were 100–500 μm (36.7%) and 500–1000 μm (33.3%). Unlike seawater MPs, no MPs smaller than 50 μm were found in the atmosphere in the present study, probably because larger MPs can easily sink (Liu et al., 2020). Small-sized MPs are distributed in high atmospheric levels and have high long-range transport potential.

Obvious weathering was observed on the surface of atmospheric MPs in the SEM images (Fig. 2). Compared to seawater MPs, more suspected microbes were observed on the surface of atmospheric MPs, and the deposition of these MPs in seawater may cause the exchange of microbial communities between seawater and the atmosphere (Audr ezet et al., 2021). Moreover, previous studies have shown that ARB and ARGs are widely present in the atmosphere, even in coarse particulate matter (PM₁₀) and fine aerosols (PM_{2.5}), and that inhalable particulate matter increases the prevalence of multidrug-resistant plasmids by promoting horizontal gene transfer (Echeverria-Palencia et al., 2017; Gandolfi et al., 2011; Jin et al., 2022; Wang et al., 2019; Xie et al., 2019; Zhu et al.,

2021). Compared to MPs in the ocean, atmospheric MPs can be directly and continuously inhaled by humans owing to continuous respiratory exposure, posing a relatively high health risk (Jenner et al., 2022; Prata, 2018). Therefore, MPs suspended in the atmosphere, especially fibrous MPs, may become potential carriers of ARGs and pathogens, promoting the spread of multidrug resistance and epidemics between different environmental compartments, which should receive more attention in the future.

3.3. Distribution patterns and potential sources of MPs in the SCS

Spatial variations were observed in the extent of seawater MP pollution in the SCS. Among the 29 locations, relatively higher MP abundances were observed at eight stations, following an order of S21 > S2 > S7 > S29 > S19 > S8 > S20 > S1, all of which were relatively close to land. The lowest MP abundances were observed at stations S3 and S12. However, higher MDII values (> 0.65) were observed at stations S14–S15, S24, and S26–27, and the station with the lowest MDII value was S3. The spatial patterns of MPs can be influenced by hydrological conditions and anthropogenic activities (Jang et al., 2020; Pan et al., 2019). Therefore, the surface currents in the SCS during the sampling period were further analyzed (Fig. 4a). The results showed that the spatial distribution patterns of seawater MPs were closely related to the South China Sea Warm Current formed by the southwest monsoon and Kuroshio intrusion (Hu et al., 2000). The high-velocity currents were loaded with MPs from Southeast Asia and other regions, and part of the debris was released along the current path, such as at stations S2, S29, and S7. The vortices conveyed and accumulated MPs, forming MP hotspots in the central (stations S19 and S20) and western SCS (station S21).

Anthropogenic activities in the surrounding countries, including urban sewage discharge, fisheries, navigation, tourism, and aquaculture, can

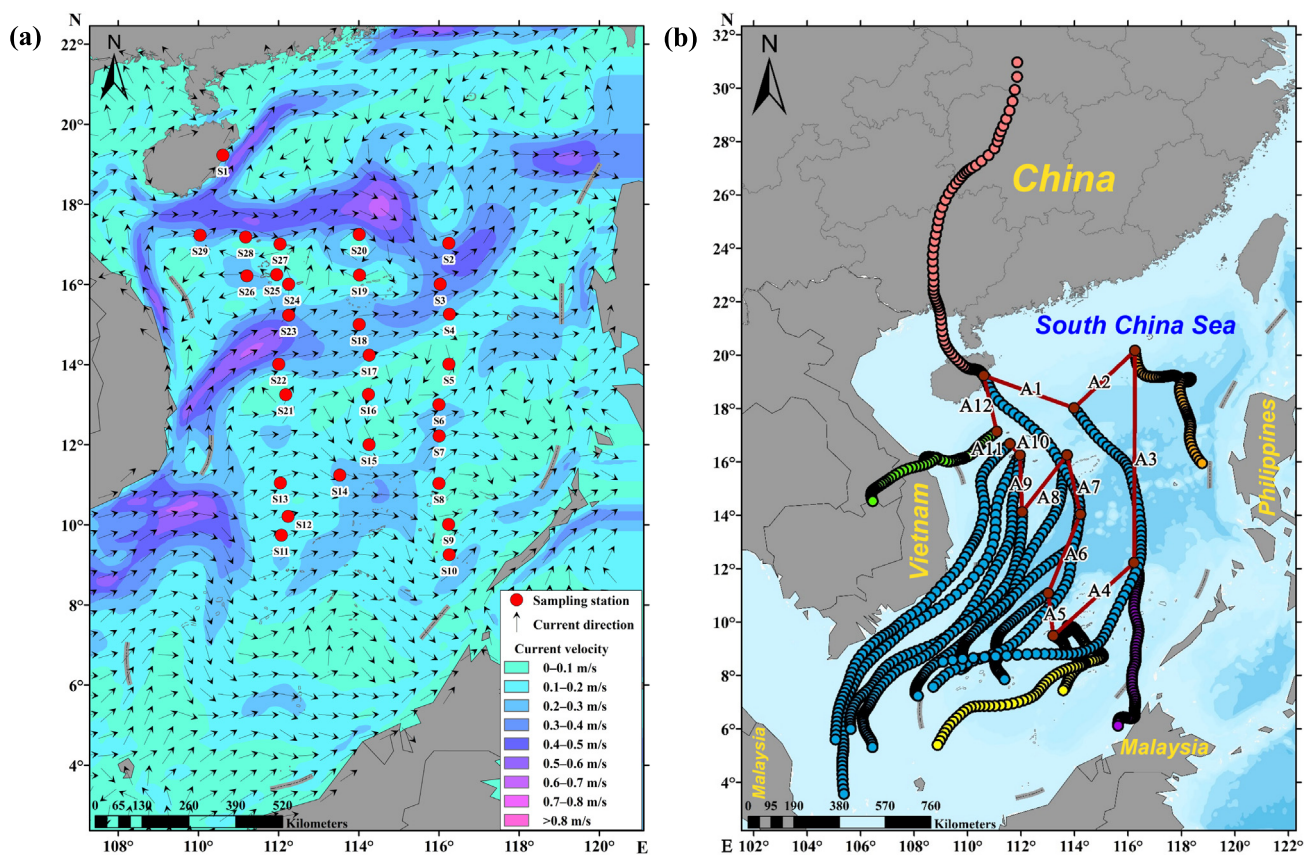


Fig. 4. Direction and strength of sea surface current at the sampling region during the period of May to June in 2021 (a) and backward trajectory of the air parcel along all sampling transects (b). The current information was obtained from the National Oceanic and Atmospheric Administration, model output HYCOM gofs3.1 hycom GLBy0.08 a0 uv3z. The dotted lines represent the three-day backward trajectories of air parcels, and solid lines represent the sampling transects.

also affect the distribution and abundance of seawater MPs in the SCS. Station S1 was located in a harbor on Hainan Island, China, and the abundant PET and PE were possibly derived from frequent industrial and transport activities, such as trade, fisheries, and navigation (Tesán Onrubia et al., 2021). As shown by the surface current results, stations S21 and S29 were located along the path of the current flowing along the coast of Vietnam; thus, the occurrence of MPs may have been affected by the direct discharge of domestic and industrial wastewater, solid waste, landfill leachate, and damaged fishing nets from Vietnam (Tran Nguyen et al., 2020). In Vietnam, 0.28–0.73 Mt. of plastic waste is discharged into the marine environment annually due to the lack of effective solutions to reduce, recycle, and dispose of plastic waste (Ngoc Anh et al., 2021; Jambeck Jenna et al., 2015). One distinctly high abundance value (station S2) was found in the eastern SCS, with the closest land being the Philippines. According to surveys, fibrous PE, PP, and PET are the dominant MPs in the Philippines and are transported by rivers, coasts, and by the direct disposal of domestic wastewater to the ocean (Esquinas et al., 2020; Galarpe et al., 2021), which may be responsible for the high proportion of fibrous PET (81.0%) at station S2. Compared to the other lands, S8 is closer to Malaysia. Owing to the lack of consistent plastic waste management and low public participation in Malaysia, approximately 0.199 trillion MP particles are emitted annually into the marine environment through sewage outlets, fisheries, aquaculture, tourism attractions, and marine parks (Praveena et al., 2018). The main MP types in the surface seawater of Malaysia are PES, PS, PA, PVC, PP, and PE (Khalik et al., 2018), which are consistent with the MP characteristics at station S8. Furthermore, the high MDII values at stations S14–S15, S24, and S26–S27 indicated that these stations had higher MP diversity and more MP sources than the other stations. These stations are located near the Xisha and Nansha islands, which are inhabited. In addition to surface currents and coastal land origins, MPs in the seawater around these islands are also derived from local sources, such as domestic wastewater and personal care products (Cai et al., 2018). The lowest MP abundance and MDII values observed at station S3 indicated that there were few other input sources.

Spatially, high MP abundance of the atmosphere was observed in seven sampling transects: A4 > A1 > A3 > A8 > A11 > A2 > A9. The lowest MP abundance was observed in A10, which was eight times lower than that in A4. The highest MDII value was observed in A9. The distribution patterns and sources of atmospheric MPs are affected by meteorological factors and anthropogenic activities (Chen et al., 2020). Southerly winds prevailed during the sampling period, driven by the East Asian summer monsoon (Fig. S3), resulting in the consistent transport of MPs in the atmosphere. Thus, the influence of meteorological factors, including pressure, temperature, wind direction, wind speed, and humidity, on atmospheric MP abundance in the SCS was determined using PCA (Fig. 5a). PC1 accounted for 68.3% of the total variance with high loadings of pressure (0.941), temperature (0.722), and abundance (0.563), whereas PC2 (24.9%) was dominated by abundance (0.800) and humidity (0.524). The discrete distances among A1–A3, A4, and A5–A12 indicate the spatial heterogeneity of the sampling stations. Atmospheric MP abundance at stations A1–A3 was predominantly influenced by pressure and temperature, whereas that at stations A5–A12 was more influenced by humidity, wind direction, and wind speed. A similar study reported that the distribution of atmospheric MPs in Shanghai was mainly determined by humidity (0.50), psychro wet bulb temperature (0.34), and barometric pressure (0.33) (Liu et al., 2019a). However, no significant correlation was observed between the MP abundance and pressure ($R = 0.441$, $P = 0.151$). In addition, MP abundance was not significantly correlated with temperature ($R = 0.123$, $P = 0.704$) or humidity ($R = -0.098$, $P = 0.762$). These results suggest that the spatial distribution of atmospheric MPs in the SCS is not affected by pressure, temperature, or humidity. Intriguingly, a significantly negative correlation was observed between atmospheric MP abundance and wind direction ($R = -0.624$, $P = 0.030$) or wind speed ($R = -0.678$, $P = 0.015$), indicating that wind or air turbulence can easily affect MP distribution owing to the low density and small size of MPs.

Backward trajectory model analysis revealed the transport pathways (except for emissions from the ocean) of atmospheric MPs (Fig. 4b). The results showed that the main sources of atmospheric MPs in the SCS were Vietnam, the Philippines, and Malaysia. According to the model simulation, the atmospheric MPs at A1 and A4–A11 may have been derived from Malaysia, those at A2 and A3 may have originated from both the Philippines and Malaysia, and those at A12 may have originated from Vietnam and China. The average abundances of atmospheric MPs derived from Malaysia (4.81 items/100 m³), the Philippines, and Malaysia (5.16 items/100 m³) were higher than those from Vietnam and China (1.77 items/100 m³), although no significant difference was observed between the contribution of atmospheric MPs from the three sources (Kruskal-Wallis test, $P = 0.368$). According to the MDII results, A9 had the highest diversity of MP composition and the largest number of MP sources. Atmospheric MPs mainly originated from air parcels passing through or being generated in Malaysia. However, there is currently a lack of surveys on atmospheric MPs in the Philippines and Malaysia, limiting the further traceability of the sources of atmospheric MPs in the SCS.

3.4. Comparison of MPs in seawater and the atmosphere in the SCS

The relationship between seawater and atmospheric MPs based on six aspects, namely, abundance, polymer, shape, color, size, and MDII, was investigated. For comparison, the sampling stations (except for S1) and transects were classified into four zones according to their geographical locations: southern SCS (SSCS, A4, A5, S7–S14), central SCS (CSCS, A6–A8, S15–S20), eastern SCS (ESCS, A1–A3, S2–S6), and western SCS (WSCS, A9–A12, S21–S29).

MPs in seawater and the atmosphere exhibited different distribution patterns in the four zones (Fig. 6a–b). The abundance of seawater MPs followed an order of WSCS (132.2 ± 142.9 items/m³) > ESCS (104 ± 119.9 items/m³) > SSCS (91.3 ± 61.7 items/m³) > CSCS (75.8 ± 44.9 items/m³). In the atmosphere, the order of MP abundance was SSCS (8.84 ± 8.13 items/100 m³) > ESCS (5.53 ± 0.99 items/100 m³) > CSCS (3.05 ± 2.35 items/100 m³) > WSCS (3.00 ± 1.55 items/100 m³). In particular, the WSCS had the highest abundance of seawater MPs and the lowest abundance of atmospheric MPs compared to the other regions. Both seawater and atmospheric MPs were highly abundant in the ESCS. The occurrence of marine and atmospheric MPs was influenced not only by the anthropogenic and environmental factors described in Section 3.3, but also by the potential interactions between the sea and air. Liu et al. (2019b) confirmed the transport of terrestrial atmospheric MPs into oceans by investigating atmospheric MPs in the western Pacific Ocean. Concurrently, Allen et al. (2020) determined that MPs can enter the marine boundary layer atmosphere through bubble burst ejection and wave action caused by strong winds or turbulence. The processes of MP deposition and emission exist simultaneously and jointly affect the migration of MPs into the atmosphere and ocean.

MPs in the seawater and atmosphere of the SCS shared similar characteristics, such as polymer type, shape, color, and size. All the polymer types (except for PC and polyisoprene) of atmospheric MPs were also identified in the seawater (Fig. 1a–b). Fibrous, granular, and film-like MPs were detected in both seawater and the atmosphere, and the predominant shape of the MPs in both seawater and atmospheric samples was fiber (Fig. 6c). Additionally, these fibrous MPs were red, black, yellow, blue, and transparent, with transparent being the most frequently detected (Fig. 6d). In the ESCS, both seawater and atmospheric MPs had the highest fibrous proportions of 88.5% and 94.7%, respectively, and this was the only zone where yellow-colored MPs were observed in both seawater and the atmosphere. Furthermore, in the CSCS and WSCS, seawater and atmospheric MPs showed the highest proportions of black and blue colors, respectively (Fig. 6c–d). According to the size distributions of MPs (Fig. 6e), MPs in the seawater and atmosphere in the CSCS shared similar size characteristics, i.e., the size category of 100–1000 μm accounted for the largest and equivalent proportion of 81.3% and 81.8% in the seawater and atmosphere, respectively, as was the case in the ESCS and SSCS. In general, a

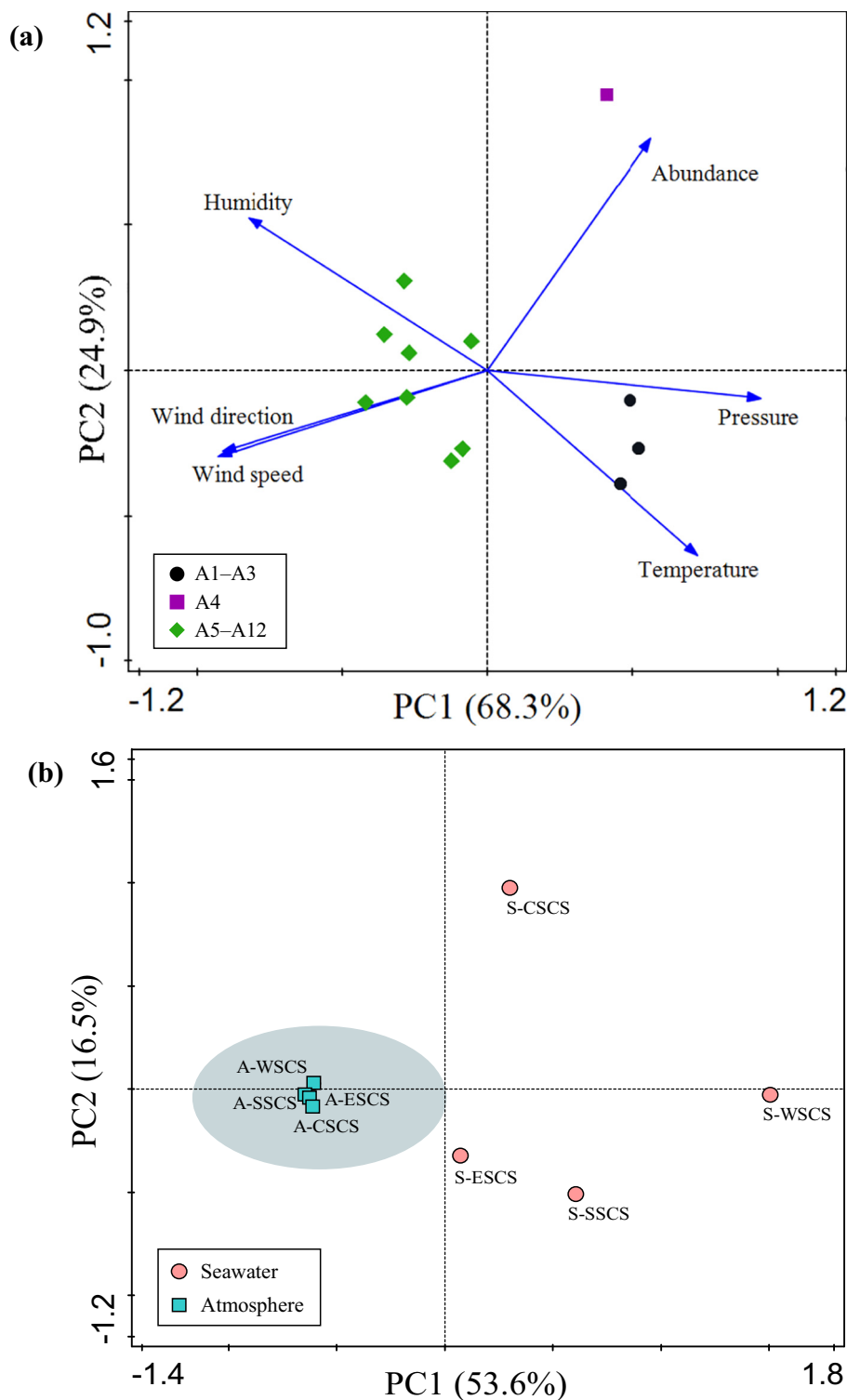


Fig. 5. Principal component analysis (PCA) plots indicate the relationship among the abundance of atmospheric MPs at the SCS and the meteorological conditions (a) and the MPs distribution patterns in seawater and atmosphere based on the polymer type, shape, color, and size of MPs (b). The label S and A refer to the seawater and atmosphere, respectively. ESCS refers to the eastern SCS (A1–A3, S2–S6), CSCS refers to the central SCS (A6–A8, S15–S20), SSCS refers to southern SCS (A4, A5, S7–S14), WSCS refers to western SCS (A9–A12, S21–S29). The vectors show the direction and strength of the environmental variable relative to the overall distribution.

small difference in size was observed between seawater and atmospheric MPs. These results could be interpreted as field-based evidence that seawater and atmospheric MPs are possible sources for each other in the same zone and highlight the interplay of continuous atmospheric MP transport, deposition, and emission from seawater (Ferrero et al., 2022).

Atmospheric MPs appear to be a “subset” of seawater MPs. In addition to having characteristics similar to those of atmospheric MPs, seawater MPs belonged to more polymer types (PP, PP-PE, PVC, PA, PAN, and PCL), shapes (fragments), colors (green and white), and size categories (< 50 μm and > 3 mm) (Fig. S2, S4). Cluster analysis based on polymer type,

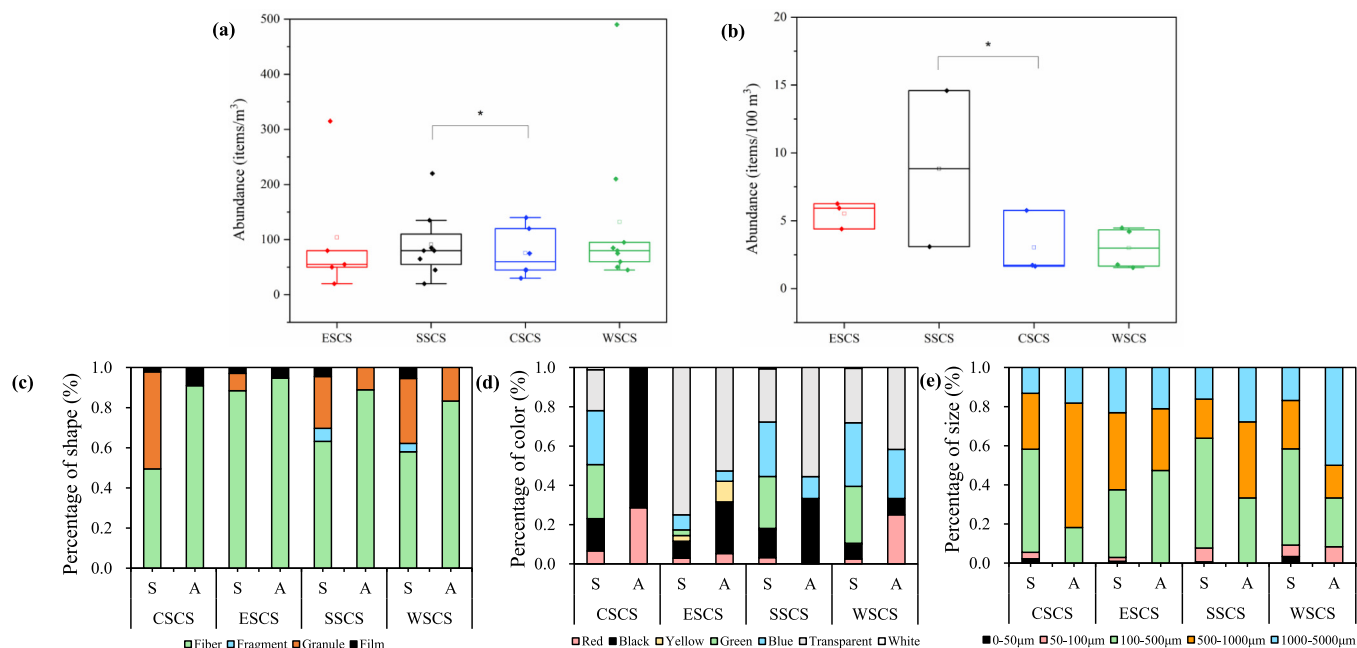


Fig. 6. Abundance (a, b), shape (c), color (d), and size (e) of MPs in seawater and atmosphere in four different regions in SCS. The label S and A refer to the seawater and atmosphere, respectively. The asterisks indicate significant difference between MP abundances in the different region ($P < 0.05$).

shape, color, and size distribution was applied to analyze the relationship between seawater and atmospheric MPs (Fig. 5b). The results showed that atmospheric MPs in the four zones shared similar polymer types and morphological characteristics and could be assigned to one group, suggesting a possible identical source of atmospheric MPs, whereas similar results were not observed for seawater MPs, illustrating the different sources of seawater MPs in the four zones. Seawater and atmospheric MPs in the same zone could not be assigned to one group, indicating that there was an apparent difference between seawater and atmospheric MPs in the same zone and that the two had different sources.

The MDII was used to reflect the compositional diversity and source complexity of MPs in seawater and the atmosphere (Fig. 7). Spatially, the MDII values of seawater MPs ranged from 0.528 to 0.690, following the order of SSCS > WSCS > CSCS > ESCS, whereas the MDII values of atmospheric MPs ranged from 0.422 to 0.523, following the order of WSCS > SSCS > CSCS > ESCS. In all four zones, the MDII values of seawater MPs

were higher than those of atmospheric MPs, particularly in the SSCS and CSCS. Overall, the average MDII value in seawater was 0.644 ± 0.078 , significantly higher than that in the atmosphere (0.472 ± 0.047) (Kruskal-Wallis test, $P = 0.029$), indicating that seawater MPs had higher compositional diversity and more complex sources than those in the atmosphere. Atmospheric MPs generally originate from emissions from urban areas and sea spray (Chen et al., 2020). Compared with the atmosphere, seawater has more abundant sources, such as rivers, runoff, wastewater, improper disposal of plastic waste in coastal environments, shipping, mariculture activities, and atmospheric dry and wet deposition (Liu et al., 2019b; Rochman, 2018). Therefore, based on these results, seawater and atmospheric MPs were deemed to be important sources of each other. Although atmospheric MPs are consistently deposited in oceans, they are not the main source of seawater MPs (Liu et al., 2019b; Wang et al., 2020b; Wang et al., 2021).

4. Conclusions

This study reports on the pollution status, patterns, sources, and interrelationships of seawater and atmospheric MPs in the SCS, which is a marginal sea surrounded by several developing countries. The results showed extensive MP contamination in the seawater and atmosphere of the SCS, which was dominated by fibrous PET. The spatial patterns of seawater MPs were mainly affected by anthropogenic activities and hydrological conditions, resulting in pollution hotspots in the ocean current vortex region adjacent to the continent. The distribution of atmospheric MPs was influenced by anthropogenic activities and meteorological factors (wind direction and wind speed), and the highest abundance was found in low-velocity southerly air parcels from Southeast Asia. There were high similarities and nonnegligible differences in the characteristics (such as polymer type, shape, color, and size distribution) of seawater and atmospheric MPs. These similarities can be considered as field evidence for the air-sea exchange of MPs. The differences between the two can be explained by the results of the cluster analysis and MP diversity integrated index analysis; that is, atmospheric MPs may be a subset of seawater MPs, which have higher diversity and more complex sources than atmospheric MPs. However, the large-scale spatiotemporal monitoring of MPs and flux quantification of MP deposition and emissions in the air-sea system should be

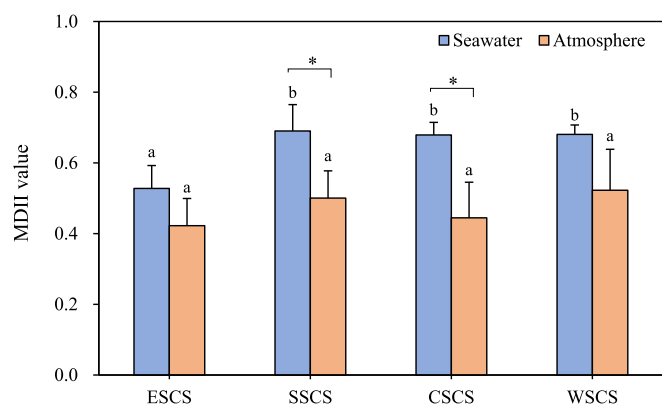


Fig. 7. MP diversity integrated index (MDII) in seawater and atmospheric samples. Error bars represent standard deviations of the means ($n = 2-9$). Different lowercase letters indicate significant difference of MDII values of seawater or atmospheric MPs among the different regions ($P < 0.05$). The asterisks indicate significant difference between the MDII values of seawater and atmospheric MPs in the same region ($P < 0.05$).

performed in the future, and the potential risks of MP-borne pathogens resulting from the exchange of MPs between surface seawater and the atmosphere should be given more attention.

CRedit authorship contribution statement

Bingjie Liu: Investigation, Methodology, Data curation, Visualization, Writing – original draft, Writing – review & editing. **Yao Lu:** Data curation, Software. **Hanqiang Deng:** Investigation. **Huanfang Huang:** Writing – review & editing, Formal analysis. **Nan Wei:** Conceptualization, Validation. **Yunlin Jiang:** Investigation. **Yuxia Jiang:** Writing – review & editing. **Liuqingqing Liu:** Writing – review & editing. **Kaifeng Sun:** Funding acquisition, Resources, Supervision, Project administration. **Hao Zheng:** Conceptualization, Visualization, Writing – review & editing.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2023.164173>.

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