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Review

Current Progress on Marine Microplastics Pollution Research: A Review on Pollution Occurrence, Detection, and Environmental Effects

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Abstract: Recently, microplastics pollution has attracted much attention in the environmental field, as researchers have found traces of microplastics in both marine and terrestrial ecological environments. Here, we reviewed and discussed the current progress on microplastics pollution in the marine environment from three main aspects including their identification and qualification methods, source and distribution, and fate and toxicity in a marine ecosystem. Microplastics in the marine environment originate from a variety of sources and distribute broadly all around the world, but their quantitative information is still lacking. Up to now, there have been no adequate and standard methods to identify and quantify the various types of microplastics, which need to be developed and unified. The fate of microplastics in the environment is particularly important as they may be transferred or accumulated in the biological chain. Meanwhile, microplastics may have a high adsorption capacity to pollutants, which is the basic research to further study their fate and joint toxicity in the environment. Therefore, all the findings are expected to fill the knowledge gaps in microplastics pollution and promote the development of relative regulations.

Keywords: microplastics; marine environment; contaminants; toxicity; adsorption

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1. Introduction

Plastics are widely used in daily life because of their excellent properties such as strong anti-corrosion ability, low electrical and thermal conductivity, high strength-to-weight ratio, and low cost to manufacture. Studies show that the production and consumption of plastics are up to 300 million tons per year [1], but plastics' recycling is not efficient due to low recovery rates and high cost [2]. Therefore, at least 10% of plastic waste enters the marine environment, which causes serious plastics pollution especially the microplastics pollution around the world [3]. In addition to the marine environment, microplastics pollution has also been found in continental waters and soil ecosystems [4–8]. Microplastics are generally referred to plastic particles smaller than 5 mm in size [9,10]. They have various colors (blue, red, black, transparent/white, etc.) and can be classified into different shape classes such as fragment, film, fiber, foam, and pellet [11,12]. The origins of microplastics include the primary and secondary sources. Primary microplastics are produced directly from the plastic production process or the pharmaceutical and personal care products (PPCPs) containing microbeads [5,13]. The secondary sources of microplastics include fragments or fibers resulting from the breakdown or weathering of plastic debris in natural environments [14,15].

The chemical properties of microplastics are relatively stable, and their degradation processes are extremely slow [16], and thus, microplastics potentially persist for a very long time in the environment [2]. Microplastics have been proved to have negative impacts on the growth of microalgae, shrimp, mussel, fish, and other organisms [10,17–22]. Moreover, the long-term aging and degradation of microplastics will release toxic additives such as antibacterial agents and plasticizers, which can also bring adverse effects to ecosystems [12,23].

Previous studies showed that microplastics have been frequently detected in the Pacific Ocean, Atlantic Ocean, and even in the Polar regions [24–28]. However, the investigation on microplastics pollution is still in the early stage so far, and data on their sources and distributions still need further enrichment. Microplastics comprise a very heterogeneous assemblage of pieces, which vary in physical properties and chemical characteristics. Thus, different approaches have been used to identify and quantify microplastics. In order to adequately estimate the composition and abundance of microplastics, it is essential to compare and assess these different approaches to develop standard evaluation methods. Up until now, considerable public attention has focused on the toxic effects of microplastics in the environment. It should be noted that more recent studies have demonstrated the ability of microplastics to carry environmental pollutants. Microplastics, as the potential contaminant vectors, may play an important role in the transport of pollutants from the aquatic environment to the biological chain, which also should be necessarily evaluated.

Therefore, the first main objective of the present work is to summarize and enrich the currently available database on the different methods for the identification and quantification of microplastics, which is expected to promote the establishment of standardized analytical methods. Secondly, the source and distribution of microplastics in the marine environment are reviewed and analyzed. Furthermore, the fate and toxicity effect of microplastics on marine organisms, together with the interaction mechanism between microplastics and environmental pollutants, have been critically evaluated. Lastly, future research directions on microplastics are pointed out. It is believed that understanding the current knowledge surrounding microplastics will provide the theoretical basis for effectively controlling plastics in order to minimize the environmental risks of microplastics.

2. Identification and Quantification of Microplastics

In order to assess the distribution and influence of microplastics in natural environments, it is essential to figure out the abundance of microplastics in these environments. However, the nature (such as size, color, surface properties, etc.) of microplastics and also the environment conditions are extremely complex. Therefore, it is important to collect and process the environmental samples through appropriate ways in order to accurately identify and quantify microplastics [29].

The commonly used sampling technologies include selection, volume capacity reduction, and bulk sampling (Figure S1). The steps for sample processing are density separation, filtration, sieving, and visual sorting. Visual sorting is one of the most commonly used methods for the identification of microplastics (using type, shape, and color as criteria). However, this process is very tedious and time consuming, and the rapid methods for sample processing are still lacking [14,30]. It also should be noted that there are still no standard procedures for sampling and processing microplastics, and this may prevent the comparison across studies.

Microplastics vary dramatically in size, shape, composition, and other physical–chemical properties; thus, the identification methods mainly focus on the chemical characterization of microplastics to identify their polymer compositions. Up to now, various methods have been applied to characterize microplastics, as shown in Table S1. Fourier-transform infrared spectroscopy (FTIR) and Raman spectroscopy are the two most common analytical methods used to identify microplastics in aquatic samples. FTIR is an appropriate method for the detection of aliphatic compounds and polyesters, and the measurement time is relatively short. However, very small particles (<10 μm) are not detectable due to the diffraction limit, and also, the spectra quality depends largely on the sample purity, particle size, and thickness [31]. Raman can identify very small particles (down to 1 μm), and it is very suitable to detect microplastics made of aliphatic, aromatic, or C=C compounds. The spectra quality of Raman depends greatly on the measurement parameters and the sample preparation methods. In order to obtain high qualitative Raman spectra, samples should be purified to avoid fluorescence, and the sample-supporting membrane filters should not show any interference to background and Raman signals during the

spectrum acquisition process [32]. Compared with FTIR, Raman takes much more time for microplastics analysis. The measurement time can be reduced but at the cost of the loss of spectra quality and the decreasing number of detectable particles. Therefore, due to the time-consuming procedures and uncertain extrapolation, the application of FTIR and Raman for reliable monitoring of microplastics is still difficult.

Pyrolysis–gas chromatography in combination with mass spectrometry (Py-GC-MS), a standard method for analyzing polymers, has been frequently used for reliable identification of the isolated microplastics particles by analyzing their thermal degradation products [33]. Py-GC-MS can also be applied for the quantitative trace analysis of microplastics on a polymer specific level based on the highly reproducible pyrolysis conditions [34]. However, it is not suited for analysis of complex environmental sample mixtures because only a single particle can be analyzed in each run [33]. In order to solve this problem, adsorption–desorption as the pretreatment method has been applied to concentrate the thermal degradation products. The sample is subjected to complete thermal decomposition with thermal gravimetric analyzer (TGA) to produce degradation products that will be adsorbed on a solid-phase adsorber [35]. Then, the adsorbed products will be analyzed by thermal desorption gas chromatography mass spectrometry (TDS-GC-MS). Compared with Py-GC-MS, TGA-TDS-GC-MS can analyze a relatively high amount of complex and not homogenous samples, which is 200 times higher than that used in Py-GC-MS. However, for both Py-GC-MS and TGA-TDS-GC-MS, the specific degradation products for the respective polymer have to be selected and analyzed first in order to identify the environmental sample accurately [33–35].

For the quantification of microplastics, the simplest and most commonly used method is counting the numbers through microscopy, which requires separating microplastics first from the obtained samples. However, this manual operation might lead to large systematic errors because of the interference of other non-plastic particles such as sea shells and minerals. Plastics always contain plastic additives such as pigments, stabilizers and plasticizers, flame retardants and so on to improve their mechanical and processing performances. A recent study quantified the microplastics through determining the concentration of additives in microplastics [36]. In that study, the alkali-assisted thermal hydrolysis was first applied to depolymerize polycarbonate (PC) and polyethylene terephthalate (PET) microplastics in a pentanol or butanol system. Then, the concentrations of the depolymerized building block compounds such as bisphenol A and p-phthalic acid were determined with LC–MS/MS to calculate the amount of microplastics. Recoveries could reach 87.2–97.1% for the PC and PET particles spiked in the landfill sludge. The method was successfully applied to determine the occurrence of PC and PET microplastics in different environmental samples such as marine sediments, indoor dust, salts, and digestive residues in organisms. In addition, this method increased the analyzing efficiency and reduced the loss of microplastics caused by picking, as the separation of microplastics from the samples was unnecessary. However, this method is not suitable to quantify the different microplastics that contain the same building block compound. Thus, the efficiency identification and accurate quantification methods for microplastics analysis still need development and improvement.

3. Source and Distribution of Microplastics

3.1. Source of Microplastics

The global evaluation of the sources for primary microplastics in marine environment is shown in Figure 1. Synthetic fiber from the textile materials in industrial laundries and households is the largest source of the primary microplastics, accounting for 35% of the overall sources [37]. These fibers are discharged into sewage water and then potentially end up in the ocean, which can reach up to thousands of particles per cubic meter [38,39]. Tire dust from abrasion while driving and city dust from abrasion of infrastructures are another two important sources of the primary microplastics, which account for 28% and 24%, respectively. In addition, the road markings (such as paint, thermoplastic, preformed polymer tape, and epoxy), marine coatings (such as paint, polyurethane, and epoxy), PPCPs,

and also plastic pellets all account for a certain proportion for the sources of microplastics. As a result of the potential environmental risks of microplastics, experts are calling for a ban on using microbeads, which is present in PPCPs [13,40]. Recently, the effluent of wastewater treatment plants (WWTPs) as the dominant origin of microplastics pollution has attracted extensive concern. All of the above-mentioned primary microplastics may enter into WWTPs, escaping the treatment processes and then being discharged into natural waters. McCormick et al. investigated a highly urbanized river in Chicago, Illinois, USA, and found that concentration of microplastics reached or exceeded the quantities in the Great Lakes and the oceans [5]. The efficiency of WWTPs for microplastics removal can reach as high as 98.41% [41]. However, because the amount of effluent in WWTPs daily is great and the treatment processes are implemented worldwide, the total amount of microplastics discharged into the environment is still significantly huge [11]. Waste disposal, surface run-off, and atmospheric fallout also contribute to the increase of microplastics.

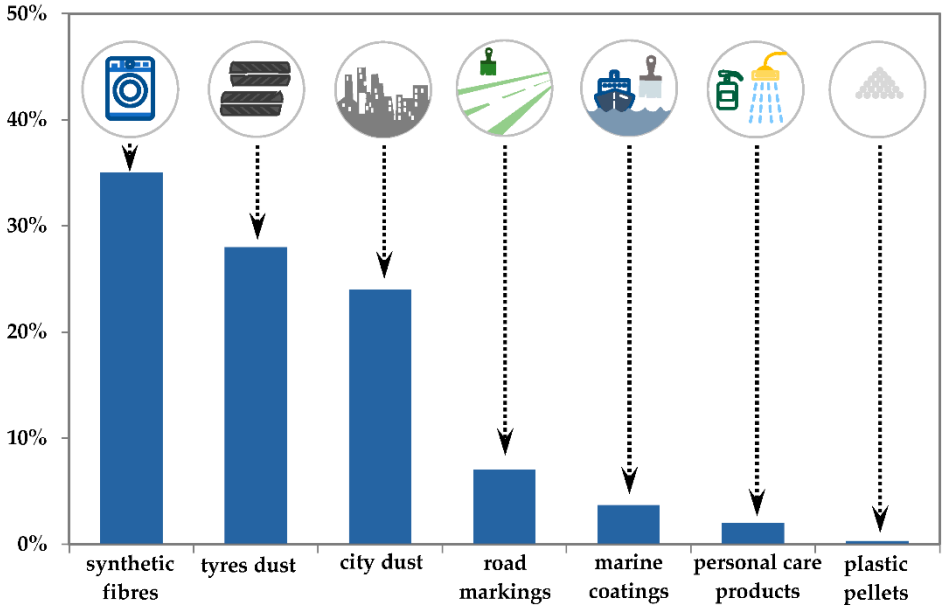

Figure 1. Global evaluation of sources for primary microplastics in the marine environment.

The decomposition of large plastic products is the dominant source of secondary microplastics in the marine environment as 80% of the marine debris is derived from land [42,43]. Plastics garbage from offshore platforms is also an important contributor. It is reported that lost or discarded fishing and entertaining polymer ropes in sailing boats can be degraded and transfer from the intertidal zone to the marine environment [44]. Plastics, especially those with chromophoric groups, may undergo photochemical reactions caused by the exposure of ultraviolet (UV) radiation. Their surfaces become weak and easy to break up, and then, they generate microplastics via friction by mechanical abrasion such as wind, waves, and sand [45]. Beaches have been considered to be the most favorable places for plastic weathering and fragmentation. Song et al. investigated the formation process of microplastics made of polypropylene (PP) and expanded polystyrene (EPS) with the treatment of 12 months of UV exposure followed by 2 months of mechanical abrasion with sand. They found that PP and EPS could produce as high as 6084 $\pm$ 1061 and 12,152 $\pm$ 3276 particles/pellet under the simulated beach environment [46]. In addition, plastic floats used in aquaculture facilities and docks can be fragmented because of the boring activity of isopods, and one adult isopod would create 4900–6300 microplastics particles during the boring process [47].

3.2. Distribution and Abundance of Microplastics

The distribution of microplastics may be affected by the change of global climate including temperature, latitude, UV intensity and so on. All cycle models show that microplastics exist in the ocean circulation, and ocean currents act as the conveyor of collection and accumulation of microplastics. Firstly, the change of the seasonal expansion and shrink of glacier will affect the flow flux of microplastics, because microplastics can be trapped by ice, and when the ice melts, microplastics can be released. Secondly, the density of most microplastics is equal to or less than the density of water, and some stronger evaporation zones will increase the density of water, which result in most of the microplastics floating on the sea and spreading to distant places with the current. The change of climate will lead to global warming; subsequently, the wind is affected, and then the wind will lead to the change of the flow of surface waters, so it is one of the reasons for changing the distribution of microplastics. Moreover, the increase of wind speed will lead to the increase of vertical mixing, which increases the amount of microplastics under a certain depth [48]. Therefore, investigating the distribution of the microplastics is significant to study the source and influence of microplastics [49].

In the recent years, microplastics have been found globally in the oceans and coastal areas, and even the deep sea and the Polar areas are not left out. Table 1 summarizes the distribution and abundance of microplastics in global regions. Levels of 0.116 particles/m^2 and 1.25 particles/m^2 were reported in the north and central–western Mediterranean Sea, respectively [26,50]. In the coastal areas of Turkey and France, the average microplastics abundance was calculated to be 0.376 particles/m^2 and 0.24 particles/m^3, respectively [51,52]. The level of microplastics abundance in San Francisco Bay was higher than other urban water bodies in North America with an average abundance of 7.0×10^5 particles/km^2 [53]. Gewert et al. reported the similar abundance of microplastics (4.2×10^5 particles/km^2) in Stockholm Archipelago, Baltic Sea [54]. In addition to the surface water, microplastics have been also detected frequently in gyres, estuaries, and sediments. For example, microplastics concentration of 13–501 particles/m^3 was observed in the North Atlantic Subtropical Gyre [25]. In the Lagoon of Venice, Italy, the density of microplastics in sediment reached as high as 2175 particles/kg dry weight [27]. In semi-enclosed bays and nearshore areas of South Korea, microplastics abundance even reached 2000 particles/m^3 [55].

It is known that a high percentage of microplastics pollution starts in inland areas and is then transported via rivers and lakes to marine systems. An average microplastics density of 2.0×10^4 particles/km^2 has been found in Lake Hovsgol, Mongolia [56]. In Lake Winnipeg, Canada, the mean concentration was about 1.93×10^5 particles/km^2 [57]. While in Subalpine Lake Garda, Italy, the mean abundance of microplastics was calculated as 75 particles/m^2 [6]. As mentioned before, WWTPs are the dominant pathway of microplastics from land to natural waters and finally into the oceans. WWTPs can be viewed as a sink for microplastics from terrestrial systems. A significantly higher abundance of microplastics in WWTPs was detected compared to that in freshwaters and the marine environment. Lares et al. reported that the microplastics concentration was 57.6×10^3 particles/m^3 in the influent of Kenkäveronniemi WWTPs located in Finland. Even after high removal treatment with advanced membrane bioreactor technology, the microplastics abundance in effluent was still as high as 1.0×10^3 particles/m^3 [58]. Murphy et al. investigated the removal of microplastics in a secondary WWTP in Glasgow, Scotland, and they estimated that 65 million microplastics would be released into the receiving water every day, even though the concentration of microplastics substantially reduced from 15.7×10^3 particles/m^3 to 0.25×10^3 particles/m^3 [11]. In Australia and Germany, the similar high abundance of microplastics in effluent of WWTPs has also been reported [59,60].

According to Table 1, the common detected types of microplastics include fragments (generated by the breakdown of larger plastic pieces), fibers, films, pellets, and foams, in which fibers and fragments are dominant for most of the research locations. In the Atlantic Ocean, 94% of microplastics were in the form of fibers [61]. Peng et al. found the similar result that fibers accounted for 93% of microplastics in sediment of Chang Jiang Estuary,

China [62]. A relatively high proportion of fibers was also detected in San Francisco Bay, Baltic Sea, and the Belgian coast [53,54,63]. In addition, fragments are another predominant composition in microplastics found in most of the current study locations. For example, fragments made up 93.2% of microplastics in the central–western Mediterranean Sea [50], and over 50% of fragments microplastics were also detected in the northeast Levantine coast, South Pacific subtropical gyre, Bay of Brest in France [28,51,52]. It can be seen from Table 1 that the most common composition of microplastics is polyethylene (PE) and PP. PE and PP are the two most commonly used plastics in both industrial and household applications such as packing, textiles, cosmetics, and so on. The chemical composition provides some possible information on the source of microplastics. For example, the high proportion of PE and PP microplastics in Stockholm Archipelago matched well with their production amount [54].

As the distribution and abundance of microplastics increases in the marine environment, the presence of microplastics in marine organisms also increases. Microplastics have been detected in various organisms from large mammals to small mollusks (Table 1). For example, Alomar and Deudero investigated the ingestion of microplastics by 125 blackmouth catsharks and found that 16.80% of the analyzed sharks ingested 0.34 $\pm$ 0.07 microplastics/individual [64]. Halstead et al. reported the occurrence of microplastics ingestion by benthic-foraging fishes in the Sydney Harbor of Australia, which was in the range of 0.2–4.6 particles/individual for the different species. It seems that microplastics abundance was relatively higher in lower trophic levels organisms, which suggested that microplastics can be transferred along food chains across various trophic levels [65]. Wild mussels *Mytilus edulis* sampled from the UK coastal waters contained microplastics of 6.4 particles/individual [66], while the number of microplastics collected in one mollusk species from the Persian Gulf of Iran reached as high as 17.7 particles/individual [67]. High microplastics pollution also occurred in cultured organisms for seafood such as oysters and sea cucumbers [68,69]. Therefore, certain sensitive organisms such as mussels are proposed as a suitable bio-indicator for microplastics pollution because of their global distribution and susceptibility to microplastics uptake [66]. The most common microplastics presented in marine organisms were fibers and fragments, which were similar to the types of microplastics in marine waters and sediments. However, the microplastics compositions were mainly CP, PET, acrylic, and rayon, which was different from that in marine environment samples. CP is widely used in food packaging and cigarette wrappers, while PET, acrylic, and rayon are always used in both textile industry and fishing gears.

Table 1. The distribution and abundance of microplastics in global regions.

Regions	Sample Method	Identification Method	Microplastics Type	Microplastics Composition	Abundance	Reference
Northwestern Mediterranean Sea	>333 μm	Microscopy	Filaments and films	PS	0.116 particles/m^2	[26]
Central–western Mediterranean Sea	>200 μm	Microscopy + ATR FTIR	Fragments (93.2%), pellets (2.2%), films (1.6%), and foams (3.1%)	PE (52%), PP (16%), PA (4.7%), PVC (2.6%), PS (2.8%), PVA (1.2%), and paints (7.7%)	1.25 particles/m^2	[50]
Northeast Levantine coast, Turkey	>333 μm	Microscopy	Fragments (60.1%), films (29.8%), filaments (7.3%), foams (2.7%), and granules (0.1%)	–	0.376 particles/m^2	[52]
Atlantic Ocean	>250 μm	Microscopy + FTIR	Fibers (94%) and fragments	PES (49%), PA or acrylic/PES (43%)	1.15 particles/m^3	[61]
Bay of Brest, France	>335 μm	Microscopy + Raman	Fragments (53%), fibers (25%), foams (11%), films (8%) and pellets (3%)	PE (67.4%), PP (16.5%) and PS (16.1%)	0.24 particles/m^3	[51]
San Francisco Bay	>333 μm	Microscopy	Central Bay: Fragment (34%), fiber (48%), pellet (1%), foam (5%) and film (1%); Southern Bay: Fragment (60%), fiber (22%), pellet (2%), foam (9%) and film (7%)	–	7.0×10^5 particles/km^2	[53]
Stockholm Archipelago, Baltic Sea	>335 μm	Microscopy + FTIR	Fibers (82%) and fragments	PE (24%), PP (53%), and PS (5%)	4.2×10^5 particles/km^2	[54]
South Pacific subtropical gyre	>333 μm	Microscopy	Fragment (79%), pellet (2%), line (14%), and film (5%)		2.69×10^4 particles/km^2	[28]
North Atlantic subtropical gyre	>10 μm	Microscopy + Raman	Fibers (40%) and particles	PE (42%), PP (6%), PS (4%), PA (11%), PU (3%), PVC (1.8%), and PES (6%)	13–501 particles/m^3	[25]
South Korea coastal areas	>20 μm	Microscopy + μ-FTIR	Fragments (81%) and fibers (18%)	PP and PE	10–2000 particles/m^3	[55]

Table 1. *Cont.*

Regions	Sample Method	Identification Method	Microplastics Type	Microplastics Composition	Abundance	Reference
Goiana Estuary, Brazil	>45 μm	Microscopy	Soft plastic (41.08%), paint chips (29.11%), hard plastic (28.42%), and threads (1.4%)	–	0.26 particles/m^3	[70]
Sediment of Chang Jiang Estuary, China	–	Microscopy + μ-FTIR	Fibers (93%), fragments (6%), and pellets (1%)	Rayon (63.1%), PES (18.5%), and acrylic (13.9%)	121 particles/kg d.w.	[62]
Sediment of Lagoon of Venice, Italy	>32 μm	μ-FTIR + ESEM-EDS	Fragments (86%), fibers (11%), films (2%), and pellets/granules (1%)	PE, PP, and PS	672–2175 particles/kg d.w.	[27]
Sediment of Bay of Brest, France	>335 μm	Microscopy + Raman	Fragments (71%), fibers (21%), and films (8%)	PE (53.3%), PP (30%), and PS (16.7%)	0.97 particles/kg d.w.	[51]
Sediment of Belgian coast	>38 μm	Microscopy + FTIR	Fibers (59%), granules (25%), films (4%), and spherules (12%)	PP, PS, nylon, PVA, and PE	390 particles/kg d.w.	[63]
Sediment of North Atlantic Ocean	>35 μm	Microscopy + μ-Raman	Particles	PE and PP	3 particles/25 cm^3	[71]
Subalpine Lake Garda, Italy	>2.2 μm	Raman	–	PE (33%), PS (33%), PP (25%), and PA (8%)	75 particles/m^2	[6]
Lake Hovsgol, Mongolia	>333 μm	Microscopy	Fragment (40%), foam (38%), line/fiber (20%), pellet (1%), and film (1%)	–	2.0×10^4 particles/km^2	[56]
Lake Winnipeg, Canada	>333 μm	Microscopy + SEM-EDS	Fibers (>90%), films, and foam	–	1.93×10^5 particles/km^2	[57]
Kenkäveronniemi WWTPs, Finland	>250 μm	Microscopy + FTIR + Raman	Fibers (82.8%) and particles (11.4%)	PES (79.1%), PE (11.4%), and PA (3.7%)	Influent: 57.6×10^3 particles/m^3 Effluent: 1.0×10^3 particles/m^3	[58]
WWTPs in Sydney, Australia	>25 μm	Microscopy + ATR-FTIR	PE, PET, Nylon, PP, PS, PVC	–	Effluent: 1.5×10^3 particles/m^3	[60]
WWTPs in Glasgow, Scotland	>65 μm	Microscopy + FTIR	Flakes (67.3%), fibers (18.5%), film (9.9%), beads (3.0%), and foam (1.3%)	PES (28%), PA (20%), PP (12%), acrylic (12%), alkyd (8%), PE (4%), PS (4%), and PET (4%)	Influent: 15.7×10^3 particles/m^3 Effluent: 0.25×10^3 particles/m^3	[11]

Table 1. *Cont.*

Regions	Sample Method	Identification Method	Microplastics Type	Microplastics Composition	Abundance	Reference
WWTPs in Lower Saxony, German	>10 µm	Microscopy + FTIR	–	PP, PE, and PA	Effluent: 9×10^3 particles/m^3	[59]
Shark *Galeus melastomus* from western Mediterranean Sea	Filed study	Microscopy + FTIR	Filaments (86.36%), fragments (12.12%), and film (1.51%)	CP (33.33%), PET (27.27%), PP (12.12%), and polyacrylate (12.12%)	0.34 ± 0.07 particles/individual	[64]
Fishes in Sydney Harbour, Australia	Filed study	Microscopy + ATR-FTIR	Fibers (83%) and granules (17%)	acrylic, PES, and rayon	0.2–4.6 particles/individual	[65]
Hymenaster pellucidus from North Atlantic Ocean	Filed study	Microscopy + ATR-FTIR	Fibers (87%) and fragments (13%)	acrylic	1.62 ± 0.9 particles/gram tissue	[72]
Shrimp *Aristeus antennatus* from northwestern Mediterranean Sea	Filed study	Microscopy + FTIR	Fibers	PET (57.1%), PA (28.6%), and rayon (14.3%)	–	[73]
Molluscs from the Persian Gulf, Iran	Filed study	Microscopy + FTIR	Microfibers (>50%), fragments (26%), films (14%), and pellets (2%)	PE, PET, and nylon	3.7–17.7 particles/individual	[67]
Mussels *Mytilus edulis* from UK	Filed study	Microscopy + µ-FTIR	Mostly fibers and a small number of fragments	PP and PES	1.1–6.4 particles/individual	[66]
Cultured oysters from China	Filed study	Microscopy + µ-FTIR	Fibers (60.67%), fragments (19.95%), films (10.26%), and pellets (9.11%)	CP (41.34%), PE (22.97%), PET(15.19%), PP (9.89%), PA (4.95%), PS (2.47%), PC (1.77%), and PVC (1.41%)	2.93 particles/individual	[69]
Cultured sea cucumbers from China	Filed study	Microscopy + µ-FTIR	Fibers, fragments, and pellets	CP	10 particles/individual	[68]

PE: polyethylene, PA: polyamides, PVC: polyvinyl chloride, PS: polystyrene, PET: polyethylene terephthalate, PVA: polyvinyl alcohol, PES: polyester, CP: cellophane.

4. Fate and Effects of Microplastics in the Marine Environment

4.1. Effects of Microplastics on Marine Organisms

According to the current field surveys, microplastics have been widely detected in various natural marine organisms. Ingestion has been widely accepted as the primary way for the marine organisms to uptake microplastics because the particles are always mistaken for food. About 15.06 particles/organism of microplastics was observed in *Daphnia magna* when exposed to 100 mg/L of PE microbeads for 21 days [74]. The ingestion of microplastics varies with the particle size, surface properties, and the age of marine organisms. Cole and Galloway found that the proportion of oysters ingesting microplastics decreased with increasing plastic size. Older oyster larvae could consume a full range of microplastics, and positive-charged microplastics were accumulated more than the virgin and negative-charged microplastics [75]. Although relative high ingestion by marine organisms, no significant negative effects were found on their normal growth, survival, and reproduction [3,76–78]. Ingested microplastics particles could be readily well egested and depurated from organisms, thus resulting in no acute biological effects [75,78].

However, more and more studies reported the negative effects of microplastics on some sensitive organisms. Prolonged exposure to PS microplastics significantly altered the feeding capacity and physically inhibited the fertilization of marine copepods, resulting in the decrease of reproductive output [79,80]. Gardon et al. investigated the impact of PS microbeads on the physiology of oysters and found a significant decrease of assimilation efficiency because of the decrease in energy gain. Oysters' gonads might provide the missing energy to maintain their metabolism from the energy balance perspective, thus producing the negative repercussions on reproduction [81]. Microplastics could also cause significant effects on marine organisms at the tissue and cellular levels. von Moos et al. reported that *Mytilus edulis* L. presented strong inflammatory response when exposed to PE microplastics. PE particles would be taken into the stomach, transported into the digestive gland, and accumulated in the lysosomal system of the mussel, which brought the formation of granulocytomas and lysosomal membrane destabilization [82]. PS microplastics were also accumulated in the zebrafish organs and then caused the inflammation and lipid accumulation in fish liver [83]. Recently, several studies have attempted to reveal the toxicity effect of microplastics at the molecular and even genetic levels. Microplastics could lead to significant adverse effects on the growth and reproduction of monogonont rotifer, with the increasing of the reactive oxygen species (ROS) production and the enzymatic activities of antioxidants. The mitogen-activated protein kinases (MAPKs) signaling pathways were significantly activated to defend against the microplastics-induced oxidative stress [84]. Similarly, a significant increase in the activities of antioxidant enzymes and decrease in the detoxifying enzymes were also observed for coral reef to acute microplastics exposure. Transcriptomic analysis revealed that 134 and 215 coral genes related with c-Jun N-terminal kinase (JNK) and extracellular regulated protein kinase (ERK) signal pathways were up-regulated and down-regulated, respectively, to repress the detoxification and immune system [85].

There are various factors influencing the toxicity of microplastics. Size-dependent negative effects of microplastics have been reported frequently. Lu et al. found that 5 μm PS accumulated in zebrafish gills, liver, and gut, while 20 μm PS only accumulated in gills and gut because the 5 μm particles could enter the circulatory system and be transferred to the liver [83]. Lee et al. investigated the two-generation chronic toxicity of different-sized PS microbeads on copepod *Tigriopus japonicus*. The results showed that the copepod mortality reached over 10% when exposed to 0.05 μm PS microbeads at 1.25 μg/mL. For 0.5 μm PS microbeads, only the highest concentration (25 μg/mL) significantly decreased the survival of the F1 generation. The 6 μm PS microbeads did not affect the survival of both F0 and F1 generations [80]. Similar size-dependent effects of PS microbeads on monogonont rotifer have also been observed. Jeong et al. found that 0.05 μm microbeads exerted the most deleterious effects on the growth rate, fecundity, life span, and reproduction time of *Brachionus koreanus*. The antioxidant-related enzymes and MAPK signaling pathways

were significantly activated in a size-dependent manner [84]. All the results revealed that smaller microplastics were more toxic, which indicated that the environmental effects of nano-sized plastics needed more research attention.

The toxicity of microplastics also depends on their surface chemistry. In laboratory studies, functionalized microbeads are always used to investigate the impact of surface properties on microplastics toxicity. Watt et al. found no significant adverse impact of both carboxylated PS (PS-COOH) and aminated PS (PS-NH_2) on gill function of shore crab *Carcinus maenas*, but their distributions in gills were quite different because of their different surface-binding capacity within the gill tissues [86]. Della Torre et al. observed that PS-COOH had no toxicity and only accumulated in the digestive tracts of sea urchin embryos, while PS-NH_2 had severe developmental defects on embryos as they caused cellular apoptosis and embryonic malformations [87]. A similar higher toxicity of PS-NH_2 on Pacific oyster larvae has been observed because PS-NH_2 beads were consumed and retained in the intestinal tract for longer than PS and PS-COOH beads [75]. Luan et al. also found that PS-NH_2 was more toxic than PS-COOH to clams at three key development stages of their life history, as the smaller particle size and positive surface charges of PS-NH_2 favored the damage of embryo membrane and the translocation through the digestive gland [88]. What is more, functional groups could enhance the combination toxicity of microplastics with other chemicals. Kim et al. found that the immobilization of *Daphnia magna* exposed to Ni combined with PS-COOH was higher than that of *Daphnia magna* exposed to Ni combined with PS [89].

In addition to the adverse physiological effects aroused from the direct contact or ingestion of the microplastics to marine organisms, microplastics, especially those containing additives, may also pose additional hazardous effects. Most additives are not chemically but physically bound to the plastic; therefore, the leaching of these additives from microplastics could be accelerated through the breakdown or swelling in natural marine environment and became available to marine organisms [90]. To investigate the potential risk, Li et al. quantified the effects of leachates from seven recyclable plastics on the survival and settlement of barnacle *Amphibalanus amphitrite*. The results revealed that leachates from plastics significantly increased the barnacle nauplii mortality and inhibited barnacle cyprids settlement on glass. A high number of chromatographic features were detected, revealing that a complex mixture of substances was released in plastic leachates, which were associated with resultant toxicity [91]. Similarly, Oliviero et al. exposed the sea urchin *Paracentrotus lividus* to the micro-sized PVC commercial products and found that the leached substances resulted in the decrease of larval length in plutei and the block of larval development in sea urchin embryos [92]. In addition to the man-made microplastics, several studies investigated the leachate toxicity associated with the natural collected microplastics. Nobre et al. studied the effects of virgin and beach-collected microplastics on the development of *Lytechinus variegatus* embryos. The results showed that the toxicity of virgin microplastics was higher than that of the beach-collected microplastics with increasing anomalous embryonic development up to 66.5%, which was attributed to the higher amount of plastics additives in virgin plastic particles [93]. However, a higher toxicity of natural collected microplastics has been also observed compared with that of the virgin microplastics. Gandara e Silva et al. found that the embryo development of brown mussels was very sensitive to the leachate from microplastics, and the beached microplastics caused 100% dead and abnormal embryos compared with 23.5% from the virgin microplastics. The authors suggested that the contaminants desorbed from the surface of beached microplastics were much more toxic than the additives leached from the virgin microplastics [94]. A recent study compared the in vitro and in vivo toxicity of leachates from North Pacific gyre plastics together with UV-treated and untreated plastics. Similarly, the North Pacific Gyre-recovered plastic leached the highest chemical estradiol equivalent and toxic equivalency compared with the untreated and UV-irradiated plastics, which significantly induced *cyp1a* mRNA in *Oryzias latipes* larvae. The results demonstrated that the weathering and desorption of adsorbed PCBs, PAHs, and other estrogenic plasticizers were responsible for the high toxic effects of natural collected microplastics [95]. Thus,

studies are still needed to explore the interaction between different kinds of contaminants with microplastics and their resulting toxicity to organisms under environmental-relevant conditions.

4.2. Toxic Effects of Microplastics on Marine Microalgae

At present, studies on the toxic effects of microplastics on marine organisms mainly focus on animals but rarely on phytoplankton, especially microalgae. Microalgae are the primary producers in the marine ecosystem and play an important role in maintaining the long-term stability of the marine environment. The obvious toxic effect of microplastics on algae is growth inhibition [96–98]. Exposed to PVC microplastics (1 μm), the growth of *Skeletonema costatum* was reported to reduce by 39.7% at 96 h under 50 mg/L treatment [99], and the inhibitory rate for *Karenia mikimotoi* was 45.8% at 24 h under 100 mg/L treatment [100]. The growth inhibition effect also increased with the decreasing of particle size [101,102]. The inhibition effect of 6 μm PS particles on the growth of *Dunaliella tertiolecta* was not significant, but it increased by 13% and 57% in the presence of 0.5 μm and 0.05 μm PS particles, respectively [103]. Along with the inhibition effects of microplastics on microalgae, the metabolic responses of algal cells were also affected obviously [96,104–106]. After 48 h exposure of PS-NH_2, the reduction of chlorophyll content and photosynthetic efficiency of the diatom *Chaetoceros neogracile* reached 24% and 13%, moreover, a significant increase of intracellular ROS was also detected [104], which might cause the lipid peroxidation. Our previous study revealed that the contents of malondialdehyde (MDA), one main lipid peroxidation product, increased even more than ten times in the presence of microplastics compared with the control [107].

Although the above toxic effects of microplastics on microalgae have received more attention, the toxicity mechanism is quite complex (Figure 2). First, as microplastics have various shapes and rough edges, it is easy to cause mechanical damage to algae cells, such as the cell wall damage and cell fragmentation [107–110], which may eventually lead to algal cells death. The mechanical damage becomes more serious with the increasing of the microplastics concentration. Zhang et al. reported that PVC could be adsorbed on the surface of algal cells and embedded into the cell wall, resulting in damage to the cell walls and membranes [99]. The mechanical damage to algal cells has also been confirmed for the other nanoparticles such as Nano-TiO_2, ZnO, and carbon nanotubes [111–113], but there is no clear evidence to prove that microplastics can enter living algal cells. However, with the environmental occurrence of nano-sized microplastics [114–117], more research is needed to verify whether microplastics can enter microalgae cells and cause intracellular mechanical damage.

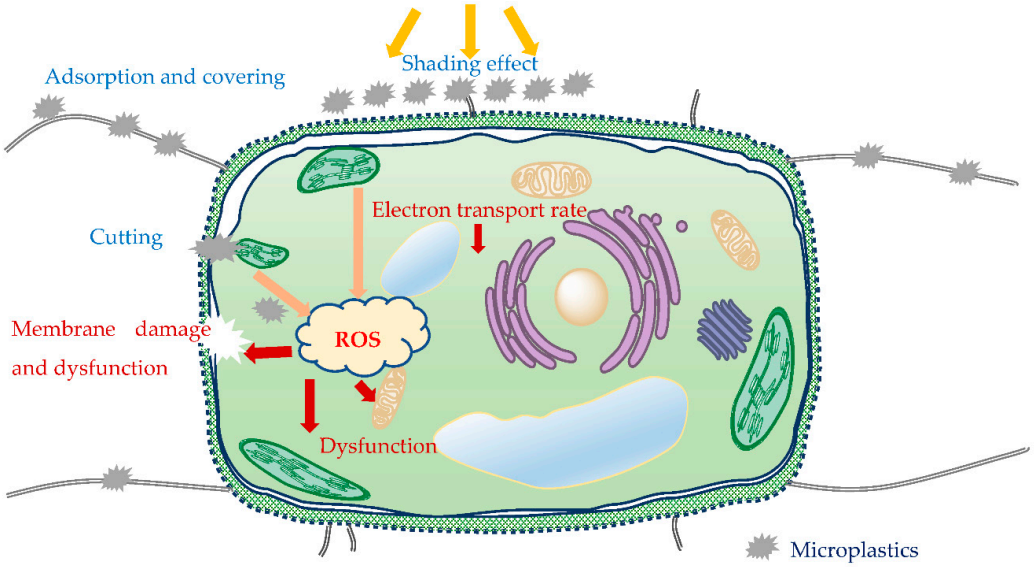

Figure 2. Schematic description of the toxicity mechanisms of microplastics to algae cell.

Apart from the mechanical damage, microplastics are expected to reduce the photosynthetic efficiency of algae through a shading effect, which is similar to other particles [97,118–120]. However, the non-contact shading experiments revealed that microplastics had no significant shading effect on the photosynthesis of microalgae [99,103]. The possible reason was that although microplastics could block some light, the rest could still meet the photosynthetic needs of algal cells as the concentration of microplastics was not high enough. However, when microplastics and algal cells contacted with each other, adsorption and hetero-aggregation may become the main reason of toxicity. First, the algal cell surface is rough with flagella, which can provide combining sites for microplastics to adhere on the algae surface [97,121–124]. The formed aggregation can not only affect the movement of microalgae but also block the absorption of light and substance exchange [97,100,125,126]. Bhattacharya et al. observed a significant decrease in CO_2 depletion in *Chlorella* because of the severe adsorption of PS on *Chlorella* cells, and harmful metabolites inside the cells could not be excreted in time, either [125]. Second, the adsorption interaction can also induce other negative effects on the structure of algal cells. For example, after 96 h exposure to 5 mg/L of 0.55 μm PS, plasmolysis, vacuolation, and distortion of the membrane structures in the *Chlorella pyrenoidosa* cells were clearly observed through TEM [108]. Furthermore, the aggregation of algal cells and microplastics may become more serious with the increasing of the exposure time, which leads to the complete covering and wrapping of algal cells and death eventually [111,112].

Otherwise, microplastics can also cause oxidative stress in algal cells, resulting in an increase of intracellular ROS content [96,105,125,127]. In plant cells, ROS mainly come from the electron transport chain in chloroplasts. When the photosynthetic efficiency decreases, the electron transport efficiency reduces, and then electrons are transferred to O_2 instead of CO_2 to form the ROS. The increase of ROS causes a lipid peroxidation reaction, which can lead to subcellular structure and cellular function damage, such as weakening the membrane selective transmission function [128,129]. It was reported that PS microplastics could induce the reduction of the photosynthetic efficiency and increase the ROS content in *Chlorella* and *Scenedesmus* cells [125]. In addition, the smaller the particle size, the stronger the oxidative stress reaction was in algal cells, thus inducing more severe lipid peroxidation in cell membrane [101,130,131]. Meanwhile, the activity of antioxidant enzymes such as superoxide dismutase (SOD) and catalase (CAT) will be enhanced to remove ROS, but algal cells will die if the ROS content exceeds their self-repair ability [96,105,128].

Moreover, as we pointed out previously, plastics additives such as plasticizers, antioxidant, flame retardants, and colorants could be leaked in the environment, which affected the toxicity of microplastics to microalgae [90,132–134]. Luo et al. found that the release amount of additives reached a maximum in 24 h, but only the high content microplastics (>1.6 g/L) had a significant inhibitory effect on the algal cell photosynthesis [134]. Normally, the additives in plastics are relatively environmentally friendly, but it has to be noted that microplastics can absorb other more toxic pollutants in the environment [97,135–140]. These chemicals can be desorbed at certain conditions, which may pose higher toxic risks to microalgae [141,142], which needs more investigation.

4.3. Trophic Transfer of Microplastics in Marine Food Webs

As a global pollution, the bioaccumulation and biomagnification of microplastics have attracted more concerns due to their potential risks and toxic effects on top predators. Thus, the trophic transfer of microplastics in marine food webs has been investigated from both field and laboratory studies. Fluorescent PS microspheres (0.5 μm) were fed through the food chain from mussels (*Mytilus edulis*) to crabs (*Carcinus maenas*), with significant detection of microspheres in the hepatopancreas of the crabs (15,033 mL^{-1} ± SE 3146 at 24 h) [143]. Setälä et al. found that 43% of the copepods and 86% of the polychaete larvae (*Marenzelleria* spp.) contained microspheres after 12 h exposure to PS microspheres (10 μm), and obvious transfer of the microspheres to higher trophic level mysid shrimps was observed after 3 h incubation with the above two mentioned zoo-

plankton species [144]. Trophic transfer of plastic fragments was also confirmed from the flying fish (*Cheilopogon rapanouiensis)* to their predator yellowfin tunas (*Thunnus albacares*) around Rapa Nui in the South Pacific subtropical gyre, even though the microplastics accumulation occurred in only a few (2%) of the tunas [145]. Microplastics transfer in marine high trophic-level taxa was also observed by *in natura* study from the prey wild-caught Atlantic mackerel (*Scomber scombrus*) to marine mammal top predator grey seals seals (*Halichoerus grypus)* [146]. Up to now, few studies involved the microplastics transfer among more than two trophic levels. Cedervall et al. studied the transport of PS nanoplastics (24 nm) through a three-level freshwater food chain from algae (*Scenedesmus* sp.) through zooplankton (*Daphnia magna*) and then to fish (*Carassius carassius*), which restrained the normal metabolization of fat reserves and severely disturbed the feeding behavior of the fish [147]. The transfer of PS nanoplastics was recently further investigated among four trophic species, including the algae (*Chlamydomonas reinhardtii*), water flea (*Daphnia magna*), secondary-consumer fish (*Oryzias sinensis*), and end-consumer fish (*Zacco temminckii).* The authors found that nanoplastics not only caused negative effects on the fish activity and induced the histopathological changes in livers but also threatened the next generation, as the plastics particles could penetrate the embryo walls [148].

After ingestion, most of the microplastics can be egested from organisms, which relieve the negative effects by microplastics in isolation [75,78]. However, the ingested microplastics can release additives and the adsorbed pollutants inside the organisms [95,149,150], which may accumulate and transfer more than the microplastics through the food web. Batel et al. investigated the transfer of microplastics and the benzo[a]pyrene through a simple artificial food chain from *Artemia* sp. Nauplii to zebrafish (*Danio rerio*) [151]. Microplastics were accumulated and transferred to fish; at the same time, benzo[a]pyrene desorbed from microplastics beads was also detected within intestinal tracts of zebrafish through direct fluorescence tracking. Diepens and Koelmans further established a generic theoretical model to simulate the transfer of microplastics and hydrophobic organic chemicals (HOCs) in food webs comprised of nine species including Atlantic cod and polar bear as the top predator [152]. The results showed that the trophic transfer of HOCs had profound effects on their biomagnification through the food chain. An interesting observation was that PCBs biomagnified less while PAHs biomagnified more when more microplastics were ingested, which was related with the metabolizable property of HOCs [152].

On the other hand, some experimental and theoretical studies predicted that ingested microplastics contaminated by pollutants would not favor chemical transfer to the organisms [153–155]. Grigorakis et al. studied the diet assimilation efficiencies (AEs) of PCBs absorbed to microplastics and food in goldfish (*Carassius auratus*) [153]. PCBs in microplastics had much lower AEs (13.36%) than that in food matrix (51.64%), and the low bioavailability of microplastics-associated PCBs suggested that microplastics were unlikely to increase HOCs biomagnification by fish in aquatic food webs. Koelmans et al. critically evaluated the possibility of the transfer of HOCs by microplastics [155]. They believed that the fraction of HOCs adsorbed by microplastics was very small compared to that adsorbed by the other marine media; therefore, microplastics ingestion was not likely to increase the exposure and risks of HOCs in the marine environment. At this time, the data on the possible transfer of contaminated microplastics through trophic levels are still lacking. More studies are needed to fulfill this gap in order to provide more evidence on the ecological risk assessment of marine microplastics.

4.4. Joint Toxicity of Microplastics with Other Chemicals

Microplastics can interact with environmental pollutants due to their large and relative hydrophobic surface, which may affect the joint toxicity of microplastics and the other chemicals from the individual level to molecular or even genetic levels. Organic contaminants such as PAHs, PCBs, and PBDEs on microplastics could be transferred into organisms [152,156], which led to the accumulation of these chemicals and much higher potential risks to marine organisms. Microplastics increased pharmaceuticals' toxicity to

marine microalgae (*Tetraselmis chuii*) with a higher inhibition of growth rate and lower chlorophyll concentration, and EC50 of doxycycline decreased almost 50% [106]. In the presence of PE microspheres (1–5 μm), the pyrene-induced fish mortality, the isocitrate dehydrogenase (IDH) activity, and acetylcholinesterase (AChE) activity were all decreased, which might increase the mortality in natural fish populations [157]. When exposed to a mixture of PE microplastics with PBDEs adsorbed from the marine environment, PBDEs concentration in *Oryzias latipes* increased significantly. Severe liver histopathology including glycogen depletion, fatty vacuolation, cellular necrosis, and lesions was also observed [158]. Exposure to microplastics and associated chemicals promoted endocrine-disrupting effects in fish, and a significant down-regulation of choriogenin, vitellogen, and estrogen receptor genes' expressions was observed [159]. The severe joint toxicity of microplastics with heavy metal contaminants has also been reported. For example, Luís et al. found that the microplastics–Cr(VI) complex greatly decreased the fish predatory behavior and inhibited the activity of AChE (31%) compared to the individual microplastics and Cr(VI) [160].

However, up to now, it is still a matter of debate whether their joint toxicity will be enhanced to organisms because the hypothetical roles of microplastics as vectors of environmental contaminants have been challenged. Herzke et al. found that persistent organic pollutants (POPs) concentrations in fulmars from Norway were not in accordance with their stomach plastic concentrations, indicating that plastic did not act as the POPs carrier to fulmars. Through calculation from a dynamic bioaccumulation model, the flux of POPs by the ingestion of natural prey was four orders of magnitude higher than the flux of POPs through plastic ingestion [154]. Even considering different conditions of pH, temperature, and gut surfactants, the predicted contribution of plastics ingestion to the overall body burdens of adsorbed organic contaminants in marine organisms was very small [161]. Both of the above modeling studies suggested that plastics ingestion was not the main way for chemicals bioaccumulation. In laboratory studies, microplastics also failed to enhance organic contaminants toxicity to organisms even under the extreme scenario conditions (microplastics concentration up to ppm level). The addition of microplastics (10 mg/L) did not increase the toxicity of 4-n-nonlphenol to both normal and starved sea-urchin larvae because sea-urchin larvae could egest microplastics after hours of ingestion without allowing significant pollutants desorption [162]. Even after ingesting relatively high doses of environmental contaminated microplastics, the rainbow trout did not show any obvious adverse hepatic stress in liver, and the fillets quality was not affected, either [163]. However, the specific properties of microplastics should be considered. For example, Kim et al. found that the joint toxicity of origin PS microplastics and Ni to *Daphnia magna* was antagonistic but synergistic for carboxyl-PS microplastics in combination with Ni [89]. Moreover, nanoplastics (50 nm) and phenanthrene had additive joint toxicity to *Daphnia magna*, while microplastics (10 μm) did not affect the bioaccumulation, dissipation, and transformation of phenanthrene [164]. These results revealed the important roles of functional groups and particle size of microplastics on their joint toxicity effect. Therefore, further studies on joint toxicity using various microplastics and contaminants are still warranted.

4.5. The Interactions between Microplastics and Contaminants

No matter whether microplastics are able to serve as an important transport vector of chemicals, there is no doubt that microplastics can act as the sinks for various contaminants in the environment, which has been found frequently from survey studies on environmental-collected microplastics. Microplastics are capable of concentrating pollutants, increasing their concentrations up to several orders of magnitude greater than those in the background. For example, high levels of PAHs, PCBs, and DDTs were detected in microplastics pellets collected from the Portuguese coast beaches, with the concentrations reaching 44,800 ng/g, 223 ng/g, and 41 ng/g, respectively [165,166]. Microplastics in the Canary Islands were reported to carry very high levels of organochlorine pesticides (up to 13,489 ng/g), as this region was once most heavily polluted by these pesticides [167].

In addition to the organic contaminants, microplastics have also been found contaminated by heavy metals. The presence of Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn, Al, Sn, Ti, and As in beach-collected microplastics have been recently reported [168–171]. The spatial pattern of pollutants in microplastics reflected the differences of these chemicals' application in different coastal regions. Furthermore, the contaminants adsorbed by microplastics still pose the potential to enter into the marine biota, although there are inverse viewpoints [106,152,156–159]. Therefore, the interactions between contaminants and microplastics are increasingly being studied to better understand the associated environmental risks.

Different types of microplastics have different affinity to environmental contaminants, which is related with the inherent properties of microplastics (Figure 3). Polymers commonly consist of crystalline regions and amorphous regions, and the crystallinity refers to the proportion of the crystalline regions. Polymer chains are closely packed in the crystalline area but loosely arranged in the amorphous area. In general, polymers with relatively lower crystallinity allow a greater diffusion of contaminants into the polymer than those with higher crystallinity. The adsorption of HOCs such as phenanthrene, lindane, and naphthalene by different types of PE were reported to be negatively correlated with the crystallinity of the polymers [172]; however, crystallinity is not the only influencing factor among the various kinds of microplastics. The adsorption of PAHs, antibiotics, phthalate esters, and perfluoroalkyl substances by the three most common PE, PS, and PVC microplastics showed very different trends, even though their crystallinity followed the order of PE > PS > PVC [139,173–177]. Plastics can be divided into the rubbery-like and glassier-like polymers according to their glass transition temperatures (*Tg*). PE has a flexible rubbery structure with greater segmental mobility and free volume at room temperature due to relatively low *Tg* of about −120 °C, which favors the diffusion and partition of pollutants into the polymer [178]. PS and PVC can be viewed as glassier polymers with high Tg values of about 100 °C and 85 °C, respectively. The presence of the benzene ring and chloride atom reduces the segmental mobility and free volume within the polymers [178]. Thus, PE commonly has a higher affinity for contaminants than the other types of plastics [174–176]. However, this generalization does not always apply to all contaminants. For example, PS was reported to have higher adsorption capacity for tylosin, tetracycline, PCBs, and other organic compounds than PE. The possible reasons were proposed to be the additional π–π interaction and polar interaction because of the benzene rings [140,177,179,180]. Thus, properties of specific microplastics and contaminants should be considered when evaluating their interactions.

There are many other different factors that may affect the adsorption behavior of microplastics. Particle size has been investigated frequently. There is no doubt that the adsorption capacity will increase with the decreasing size of microplastics [181–185]. It should be noted here that nanoplastics are receiving more concerns. Up to now, information on the environmental loads of nanoplastics is still not available. Release from products and the degradation/fragment of larger plastics should be the two most important sources of nanoplastics [186]. During the limited studies on the potential environmental impacts of nanoplastics, nano-PS had almost two orders higher adsorption capacity to PCBs than micro-PE [180]. Nano-PS was also reported to enhance the transport and spread of organic contaminants as an important carrier [187]. Therefore, there is a need for more research to further understand the interactions between nanoplastics and environmental contaminants.

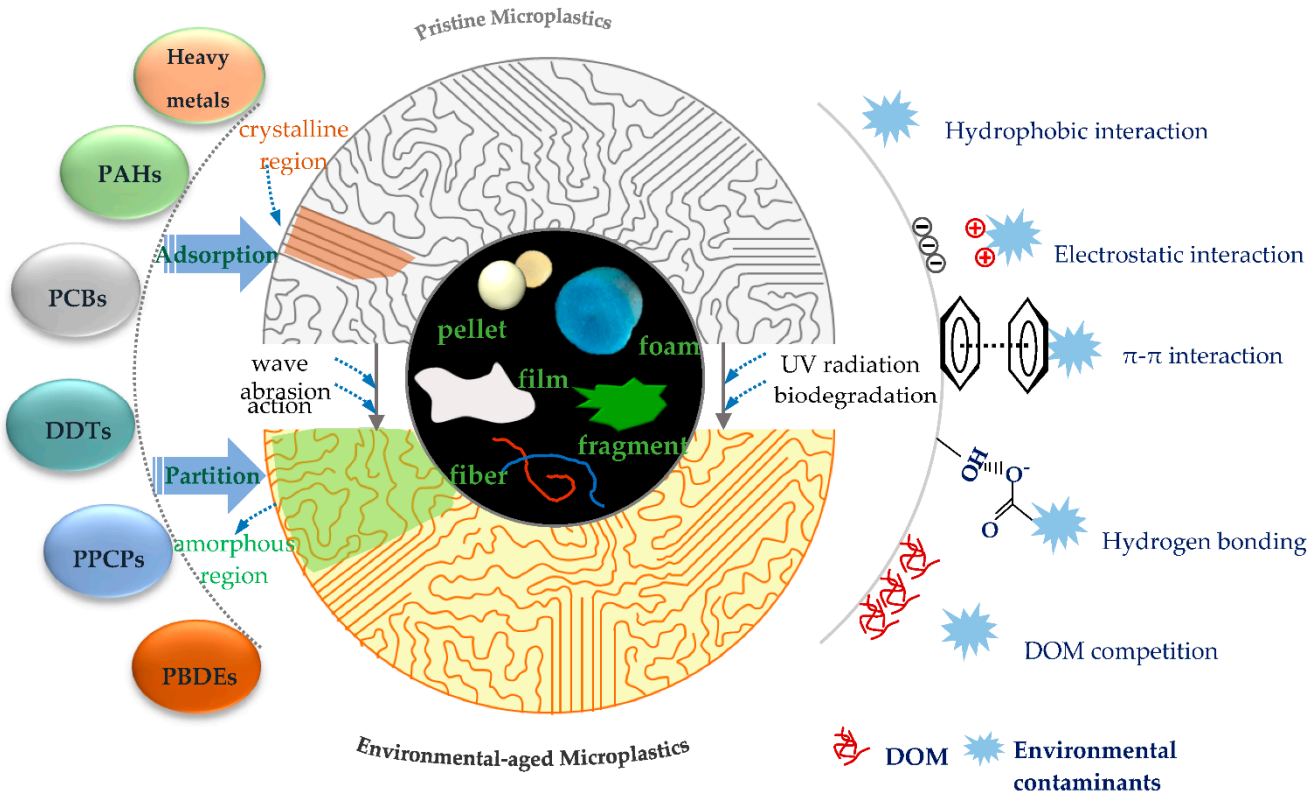

Figure 3. Schematic description of the interaction and mechanisms between environmental contaminants with microplastics.

Once released into environment, plastics will undergo the multiple weathering processes to form the aged microplastics due to the mechanical abrasion, wave action, UV radiation, biodegradation and so on [188]. High levels of organic and metal pollutants on environmental-collected microplastics proved the important role of these aged microplastics as contaminants sink [165–171]. During the weathering process, wrinkles and cracks will be formed on microplastics, increasing the surface roughness and surface area [189], which allows pollutants to effectively diffuse and adsorb in microplastics through the fine cracks [190]. A large amount of oxygen-containing functional groups (e.g., carbonyl and hydroxyl groups) are also introduced on aged microplastics [190], which may increase the polarity and the adsorption affinity for hydrophilic compounds and heavy metals [189,191]. However, a lower adsorption of hydrophobic pollutants on aged microplastics was also reported compared with the pristine particles [192]. Liu et al. investigated the aging behaviors of microplastics under simulated advanced oxidation conditions and found that the aging degree and properties of microplastics closely related with the adsorption behaviors [193]. We note that the environmental behaviors of aged microplastics that resulted from biofouling, microbial degradation, and other weathering processes are rarely studied. Upon the release of microplastics to the water and/or sediment environment, biofilms quickly form on their surfaces (taking place in minutes to hours), which can influence the adsorption processes of microplastics [194]. Biofilms can act as a potential adsorptive phase because of the presence of extracellular polymeric substances such as polysaccharides, proteins, lipids, and other biopolymers. For example, Johansen et al. found that biofilms enhanced the adsorption of strong and weak cations (^{137}Cs and ^{90}Sr) onto microplastics, revealing that microplastics could be viewed as a potential sink for the environmental radiotracers [195]. However, biofilms can also act a barrier for the diffusive uptake and release of chemicals by increasing the resistance for mass transfer into and out of the microplastics [194]. Therefore, more knowledge on the multiple weathering factors on the properties of microplastics and their resulting adsorption interactions with pollutants still need further clarification.

In addition to the above influencing factors, the adsorption of contaminants by microplastics is highly affected by environmental aquatic chemistry, which has been studied a lot. Normally, pH has no significant effect on the adsorption of weak/non-polar hydrophobic compounds by microplastics [173,174,196]. However, the adsorption of hydrophilic contaminants by microplastics may change [140,174,179,197], because the solubility and dissociation of these chemicals is pH-dependent, which will influence the hydrophobic and electrostatic interactions with microplastics. Salinity can also affect the adsorption behavior of microplastics but with different results. Compared with that of freshwater, the adsorption capacity in seawater was enhanced for triclosan, perfluorooctanesulfonate, phthalate esters, and PCBs by microplastics [173,174,180,198], which might be due to the salting-out effect as the solubility of these weak/non-polar contaminants was decreased. However, a decreased adsorption capacity of microplastics with increasing the solution salinity to a certain extent was also reported for some hydrophilic compounds (e.g., antibiotics) [179,184,189,199]. The competition of cations for the adsorption sites on microplastics was reported to be the main reason, which reduced the electrostatic or H-binding interactions between organic contaminants and microplastics. Considering the above contradictory results, no significant effects of salinity on the adsorption capacity of microplastics have also been reported [140,185,196,198,200,201]. It seems that the influence of salinity highly depends on the properties of contaminants, which merits deep investigations. Similar to pH and salinity, the ubiquitous dissolved organic matter (DOM) presented similar effects on the adsorption capacity of microplastics. Commonly, DOM has adverse effects on the adsorption capacity of microplastics [140,180,196,198,201,202]. DOM could interact with microplastics via π–π conjugation, carboxyl groups, and C=O bonds to form a conjugated co-polymer with an elevated electron density [203], which decreased the adsorption of other organic compounds through competition for the adsorption sites. It should be noted that the adsorbed DOM might also interact with pollutants, which may counteract or even exceed the negative effects. For example, humic acid had no significant effects on the adsorption of sulfamethoxazole and phthalate esters by microplastics [173,200], while Zhang et al. observed that humic acid promoted the adsorption of oxytetracycline by microplastics through enhancing the electrostatic interaction [197]. Thereby, the role of DOM on the interactions between contaminants and microplastics needs more attention.

The desorption of chemicals from microplastics is critically important for assessing the role of microplastics as transport vector of contaminants. The reversible desorption of contaminants from microplastics implies the potential release of these chemicals once uptaken by organisms, while irreversible desorption (or desorption hysteresis) will make microplastics as the pollutant sinks. Desorption hysteresis more likely occurs on glassy microplastics than rubbery microplastics. As stated before, the rubbery domains were flexible and highly accessible for chemicals, while the molecular chain segments in glassy domains were more condensed and crosslinked. The adsorption in glassy polymers was affected by both partition and pore filling, and the latter was responsible for desorption hysteresis. Thus, Zuo et al. observed significant desorption hysteresis of phenanthrene from glassy PS but no hysteresis on 100% rubbery biodegradable poly(butylene adipate co-terephtalate) microplastics [204]. Liu et al. found that PS nanoparticles enhanced the transport of weak/non-polar chemicals in saturated soil but had no effects on polar compounds, and they proved that the desorption hysteresis of weak/non-polar chemicals from the dense and glassy PS was the main reason [187]. Due to the potential contribution of ingested microplastics to the transfer of contaminants into organisms, the desorption of chemicals in simulated gut conditions has also been studied. Bakir et al. reported that the desorption of POPs was enhanced by the gut surfactant, which reached up to 30 times higher than that in seawater [205]. A similar high and fast desorption of PCBs in artificial gut solution was also observed, but the steady-state bioaccumulation factor was predicted to decrease with the increasing ingestion of microplastics, which suggested that the ingested microplastics might play a dilution role for contaminants. However, the authors also pointed out that the desorption of plastic additives might increase the potential

accumulation of these chemicals [206]. The latest study conducted by Coffin reported that the known butylbenzyl phthalate desorbed in fish gut condition was 1.3 times higher than that in fish control, but the increase in biological estrogenicity by gut condition was 5.1 times higher than the control, which also proved that the contribution was significant by the desorbed uncharacterized plastic additives [149].

5. Conclusions

In recent years, microplastics as emerging contaminants have attracted much public attention. This review article summarized the current status of microplastics in the marine environment. The sampling methods were first described, and the identification methods including FTIR, Raman, pyrolysis/TGA-GC-MS, and other novel ways were compared to provide the researchers with the proper one to apply. Through reviewing the literature on survey studies, microplastics were found to be globally distributed in surface water, depth water, sediment, and even the Polar regions. The abundance of microplastics in WWTPs was further analyzed, indicating the important contribution of WWTPs for marine microplastics. What is more, microplastics have also been detected in field-caught and cultured marine organisms, which may bring potential risks to human. Then, the effects of microplastics on marine animals and microalgae, the trophic transfer of microplastics in marine food webs, and the joint toxicity of microplastics were assessed. Microplastics posed negative effects on marine organisms from the individual level to cellular level and even to the genetic level. The food chain transfers of microplastics and their joint toxicity were also observed even though some studies found that certain organisms might egest microplastics out after ingestion. Finally, the interactions between microplastics and environmental contaminants were systematically explored, which were controlled by the inherent properties of microplastics, nature of chemicals, and the environmental chemistry factors. Although the current studies on marine microplastics are developing quickly, research on the above aspects is still at the preliminary stage. For better understanding the environmental fate of microplastics, the following works should be addressed in the future:

(1) The current methods for the sampling and identification of microplastics need to be standardized. Efficient and adequate methods should be developed for the in-situ detection of microplastics.
(2) Although we have gained some information on the distribution and abundance of microplastics, it is still not sufficient for the global regions. More survey studies are still needed to enrich the database of microplastics pollution.
(3) As an important source for marine microplastics, investigation on the terrestrial pollution is not enough, especially for the WWTPs that we pointed out previously. Thus, the fate and transport of microplastics in WWTPs needs further study, and the microplastics-targeted treatment methods urgently need to be developed for reducing the amount of microplastics released from WWTPs to the environment.
(4) There are not yet adequate studies on the impact of microplastics to microalgae, the marine environmental producer, which still need more research from the population level to the genetic level. In addition to the low trophic level organisms, the potential transfer of microplastics and the related contaminants from seafood products to human should also be carefully evaluated.
(5) Considering the role of microplastics as vectors to transport pollutants, the chemicals adsorbed on environmentally collected microplastics should be analyzed to explore the formation of the microplastics–contaminants complex. Systematic studies are also needed to clarify the adsorption and desorption mechanisms of various chemicals on microplastics.

Supplementary Materials: The following are available online at https://www.mdpi.com/article/10.3390/w13121713/s1, Figure S1: Sampling technologies and methods of processing, Table S1: Advantages and disadvantages of the identification and quantification methods for microplastics.

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