Interaction of Microplastics and Organic Pollutants: Quantification, Environmental Fates, and Ecological Consequences

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Contents

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Abstract Microplastics which act as vectors for organic pollutant transport in environment have raised increasing concerns recently. This paper provides an overview on the interaction of plastic debris or microplastics with these organic chemicals and its effects on biological receptors. Plastic additives represented one of the most important organic pollutants associated with microplastics; the types, quantification, and migration from the plastic debris or microplastics are addressed here. In addition to the chemical additives, microplastics also adsorbed hydrophobic or hydrophilic organic pollutants from the environments due to their high surface areas and affinity for these pollutants. The mechanisms of microplastic adsorption for PAHs, PCBs, and pharmaceutics and the role of microplastic surface and solution chemistry were well discussed in the paper. The sorption affinity changed by the aging of microplastic surface was of concern in particular. The organic pollutants in the microplastics may cause toxic effects on biotas by releasing into the leachate or by contact exposure directly through microplastics ingestion. Here we reviewed the latest reports on the organic pollutant assay for the leachates from the environmental microplastics and their toxic effects on freshwater species *Daphnia magna*, brown mussel (*Perna perna*), barnacle, and microalgae using different endpoints. Bioaccumulation of organic pollutants and biological toxicology through the vector effects of microplastics were also reviewed in the paper. However, large uncertainties existed among the different studies with respect to the toxic effects of co-exposure with organic pollutants and microplastics. Therefore, further researches are recommended to be done regarding the combined effects of organic pollutants and microplastics under the different exposure scenarios.

Keywords Combined effect, Microplastics, Organic pollutant, Plastic additive, Sorption and desorption

1 Introduction

Microplastics (plastics <5 mm, including nanoplastics which are <0.1 μm) originate from the fragmentation of large plastic litter or from direct environmental emission. Their potential impacts in terrestrial ecosystems remain largely unexplored despite numerous reported effects on marine organisms [1]. Meanwhile, microplastics present in the terrestrial environment carry organic chemicals of smaller molecular size. These chemicals can penetrate into cells, chemically interact with biologically important molecules, and may disrupt the endocrine system [2]. Such organic chemicals are categorized into two groups: (1) additives, monomers, and oligomers of the component molecules of the plastics and (2) hydrophobic or hydrophilic organic compounds that are adsorbed from the surrounding environment through different mechanisms. Many of the contaminants addressed herein have known biological consequences. Furthermore, microplastics were assumed to serve as

vectors for transport of organic chemicals from environment biota and even across cell membranes to elevate intracellular stress [3]. Therefore, the objective of this paper is to review the interaction of plastic debris or microplastics with these organic chemicals and its effects on biological receptors.

2 Quantification and Environmental Fate of Plastic Additives in the Plastic Debris or Microplastics

2.1 Chemical Additives Use and Application in the Plastic Productions

Plastic additives are mainly used as plasticizers, flame retardants, stabilizers, antioxidants, and pigments, which are added to improve the performance and functionality of a plastic product. All the additives can be mainly divided into the following four categories based on their functional properties: (1) functional additives (stabilizers, antistatic agents, flame retardants, plasticizers, lubricants, slip agents, curing agents, foaming agents, biocides, etc.), (2) colorants (pigments, soluble azo colorants, etc.), (3) fillers (mica, talc, kaolin, clay, calcium carbonate, barium sulfate), and (4) reinforcements (e.g. glass fibers, carbon fibers).

Plasticizers are most commonly used for improving the flexibility, durability, and stretchability of polymeric films and reducing melt flow. Phthalate esters (PAEs) are the most commonly used plasticizers and include flexible vinyl, which is mainly added in PVC production, which can contain 10–60% phthalates by weight [4]. As they are not part of the chain of chemical compounds (polymers) of plastics, they can be released from these products. These plasticizers are used in various fields such as foods, toy manufacturing, electronics, pesticides, glues, paint solvents, personal care products, and pharmaceuticals [5].

Antioxidants are used as additives in many synthetic polymers including polyolefins (mainly PE and PP) which represent 60% of global demand for antioxidant additives. Arylamines are the most commonly used antioxidants in plastic food packaging. Phenolics and organophosphates (used to reduce hydroperoxides formed during oxidation to alcohols) are also used as antioxidants. Bisphenol A (BPA) and nonylphenols (NP) can also be used as an antioxidant or as a plasticizer in the PP, PE, and PVC polymers. Over three million tons of BPA was produced annually for the using in plastics [6].

Heat stabilizers are responsible for preventing thermal degradation of polymers when exposed to elevated temperatures, i.e., during the thermal processing of foods. Certain types of polymers, i.e., PVC, PVDC, vinyl chloride copolymers (e.g., vinyl chloride/vinyl acetate), and PVC blends require the addition of heat stabilizers in order to maintain their functionality. Organic chemicals such as alkyl organophosphates, epoxy compounds, and beta-diketones are the main types of secondary heat stabilizers.

Brominated flame retardants (BFRs) are a class of additives used in plastic products to reduce flammability. These BFRs are used in a variety of consumer products ranging from electronic devices to insulation foams. BFRs include a wide range of chemicals, of which polybrominated diphenyl ethers (PBDE), hexabromocyclododecane, and tetrabromobisphenol A represent the main BFRs used in the plastic industry [4]. Hexabromocyclododecane (HBCD) is the main additive chemical applied to polystyrene (PS) products, including EPS, and is especially prevalent in construction materials and electronic housings [4]. The global consumption of HBCD increased from 16,700 tons to 31,000 in 2001–2011. HBCD is not covalently bound to the polymer and can therefore be easily released from plastic products into the environment [7]. Organophosphate esters (OPEs), and in particular the triesters, are produced in high volumes and have been used as flame retardants and plasticizers for decades. The usage even increases due to the ban or restriction of brominated diphenyl ethers as flame retardants [8]. In 2011, 500,000 tons of OPEs were consumed globally, and annual consumption reached 680,000 tons by 2015 [9]. The OPEs can volatilize from a product or be lost through abrasion or dissolution, and much attention has been paid to the release of OPEs into the environment from products.

2.2 *Occurrence of Chemical Additives in the Environmental Plastic Debris or Microplastics*

To date, only a few studies have focused on the detection of plastic additives from MP collected in coastal beaches [7, 10–15]. A dataset summary regarding plastic additive concentrations in the plastic debris and microplastics is shown in Table 1. Mato et al. [11] detected nonylphenols in PP pellets deployed in Tokyo Bay and suggested that these compounds came from plastic additives found in the PP pellets themselves. Hirai et al. [13] observed high levels of PBDEs, BPA and nonylphenols in PE and PP fragments collected on remote or urban beaches. The source of these organic compounds was assumed to be connected with plastic additives used for the manufacture of PP and PE. A wide range of plastic additives were also identified using Pyrolysis-GC/MS with thermal desorption in MP collected from sediment of Norderney Island [10]. The identified PE, PP, PS, and polyamide-6 MP particles were associated with antioxidant additives, e.g., DEHP, DnBP, diisobutyl phthalate (DiBP), and 2,4-di-tert-butylphenol (2,4-DTBP). Moreover, Rani et al. [14] detected multiple plastic additives in plastic marine debris found on a beach in Geoje, South Korea. Indeed, the authors found BPA and phthalates in PP and PE plastic marine debris as well as antioxidants including Irganox 76 and 2,4-DTBP in PP and PE plastic marine debris. In another study which focused on ultraviolet stabilizers (UVSs) and antioxidants in the plastic debris of the beaches, it reported that

Table 1 Summary of the chemical additives measured in plastic debris or microplastic samples

Chemical additives	Sampling locations	Plastic types	Particle size	Concentration (ng/g)	References
Hexabromocyclododecane (HBCD)	South Korean coasts	EPS bead[a]	0.99 ± 0.06 mm	70,300 ± 77,800	[10]
	South Korean coasts	EPS bead[b]	0.99 ± 0.06 mm	$5.98 \times 10^6 \pm 2.12 \times 10^6$	
	South Korean coasts	Pre-expanded spherule[a]	3.22 ± 0.21 mm	5,810 ± 680	
	South Korean coasts	Pre-expanded spherule[b]	3.22 ± 0.21 mm	$2.37 \times 10^6 \pm 8.15 \times 10^4$	
	South Korean coasts	EPS product[a]	n.a.	5,380 ± 1,360	
	South Korean coasts	EPS microplastics	2–3 mm	$1.07 \times 10^6 \pm 4.50 \times 10^5$	
	South Korean coasts	Small-sized buoy (40–80 L)	Diameter 35 × 45 cm	$1.45 \times 10^5 \pm 3.79 \times 10^5$	
	South Korean coasts	Large-sized buoy (>200 L)	Diameter 50 × 90 cm	$7.11 \times 10^5 \pm 1.25 \times 10^6$	
	South Korean coasts	Recycled buoy (inside)	n.a.	$1.94 \times 10^6 \pm 8.89 \times 10^5$	
	South Korean coasts	Recycled buoy (outside)	n.a.	$7.51 \times 10^5 \pm 9.69 \times 10^5$	
	Hong Kong	Low-density EPS debris	n.a.	920 ± 1,460	
	Singapore	Low-density EPS debris	n.a.	170 ± 80	
	Brunei	Low-density EPS debris	n.a.	230 ± 140	
	Vietnam	Low-density EPS debris	n.a.	100 ± 60	
	Thailand	Low-density EPS debris	n.a.	3,510 ± 4,890	
	Taiwan	Low-density EPS debris	n.a.	2,140 ± 4,120	
	Bangladesh	Low-density EPS debris	n.a.	7,430 ± 14,300	
	Sri Lanka	Low-density EPS debris	n.a.	50 ± 20	
	Japan	Low-density EPS debris	n.a.	$1.08 \times 10^5 \pm 1.93 \times 10^5$	
	Peru	Low-density EPS debris	n.a.	$3.2 \times 10^6 \pm 2.8 \times 10^6$	
	Canada	Low-density EPS debris	n.a.	$1.47 \times 10^6 \pm 2.27 \times 10^6$	
	USA (Hawaii)	Low-density EPS debris	n.a.	4,940 ± 5,710	
	USA (California)	Low-density EPS debris	n.a.	$1.21 \times 10^6 \pm 1.98 \times 10^4$	
	USA (Alaska)	EPS fragment	n.a.	$3.35 \times 10^6 \pm 5.86 \times 10^6$	

(continued)

Table 1 (continued)

Chemical additives	Sampling locations	Plastic types	Particle size	Concentration (ng/g)	References
PBDEs	Marbella Beach, Costa Rica	PE fragments	A few mm to few cm	0.4–5.8	[13]
	Marbella Beach, Costa Rica	PP fragments	A few mm to few cm	1.6–180	
	Seal Beach, USA	PE fragments	A few mm to few cm	12.6–41.4	
	Seal Beach, USA	PP fragments	A few mm to few cm	5.3–25.5	
	Odaiba, Japan	PE fragments	A few mm to few cm	1.1–15.5	
	Odaiba, Japan	PP fragments	A few mm to few cm	1.4–2.7	
	Kugenuma, Japan	PE fragments	A few mm to few cm	0.02–1.7	
	Kugenuma, Japan	PP fragments	A few mm to few cm	3.9–230	
	Tonkin Bay, Vietnam	PE fragments	A few mm to few cm	2.1–6.5	
	Tonkin Bay, Vietnam	PP fragments	A few mm to few cm	0.3–412	
Nonylphenols (NP)	Kasai Seaside Park in Japan	Resin pellets	0.1–0.5 cm	16	[11]
	Keihin Canal in Tokyo Bay	Resin pellets	0.1–0.5 cm	8.9	
	Kugenuma Beach	Resin pellets	0.1–0.5 cm	12	
	Shiroda Beach	Resin pellets	0.1–0.5 cm	0.13	

	Marbella Beach, Costa Rica	PE fragments	A few mm to few cm	2.4–33.4	[13]
	Marbella Beach, Costa Rica	PP fragments	A few mm to few cm	5.3–3,936	
	Seal Beach, USA	PE fragments	A few mm to few cm	0–1.1	
	Seal Beach, USA	PP fragments	A few mm to few cm	7.2–130	
	Odaiba, Japan	PE fragments	A few mm to few cm	0.3–13.4	
	Odaiba, Japan	PP fragments	A few mm to few cm	8.5–22.4	
	Kugenuma, Japan	PE fragments	A few mm to few cm	1.3–706	
	Kugenuma, Japan	PP fragments	A few mm to few cm	305–1,244	
	Tonkin Bay, Vietnam	PE fragments	A few mm to few cm	0–0.7	
	Tonkin Bay, Vietnam	PP fragments	A few mm to few cm	0–551	
Octylphenol (OP)	Marbella Beach, Costa Rica	PE fragments	A few mm to few cm	0–1.6	[13]
	Marbella Beach, Costa Rica	PP fragments	A few mm to few cm	0.2–14.4	
	Seal Beach, USA	PE fragments	A few mm to few cm	0–16.8	
	Seal Beach, USA	PP fragments	A few mm to few cm	0.1–4.9	
	Odaiba, Japan	PE fragments	A few mm to few cm	0–17.4	

(continued)

Table 1 (continued)

Chemical additives	Sampling locations	Plastic types	Particle size	Concentration (ng/g)	References
	Odaiba, Japan	PP fragments	A few mm to few cm	0.2–2.6	
	Kugenuma, Japan	PE fragments	A few mm to few cm	0–49.2	
	Kugenuma, Japan	PP fragments	A few mm to few cm	0.8–27.6	
	Tonkin Bay, Vietnam	PE fragments	A few mm to few cm	0.8–1.5	
	Tonkin Bay, Vietnam	PP fragments	A few mm to few cm	0–154	
BPA	Marbella Beach, Costa Rica	PE fragments	A few mm to few cm	0–729.7	[13]
	Marbella Beach, Costa Rica	PP fragments	A few mm to few cm	0.4–35.4	
	Seal Beach, USA	PE fragments	A few mm to few cm	0–6.8	
	Seal Beach, USA	PP fragments	A few mm to few cm	0–26.2	
	Odaiba, Japan	PE fragments	A few mm to few cm	0–1.4	
	Odaiba, Japan	PP fragments	A few mm to few cm	0–2.8	
	Kugenuma, Japan	PE fragments	A few mm to few cm	0–0.5	
	Kugenuma, Japan	PP fragments	A few mm to few cm	0–3.4	
	Tonkin Bay, Vietnam	PE fragments	A few mm to few cm	0	

	Tonkin Bay, Vietnam	PP fragments	A few mm to few cm	0–263.6	
Irganox 1076	Geoje coast, South Korea	Plastics debris	n.a.	$1.06 \times 10^5 \pm 2.89 \times 10^5$	[14]
Irganox 1076	Geoje coast, South Korea	New plastics	n.a.	$1.50 \times 10^5 \pm 4.40 \times 10^5$	
Irganox 1010	Geoje coast, South Korea	Plastics debris	n.a.	$3.4 \times 10^4 \pm 4.9 \times 10^4$	
Irganox 1010	Geoje coast, South Korea	New plastics	n.a.	$9.5 \times 10^4 \pm 1.52 \times 10^5$	
2,4-DTBP	Geoje coast, South Korea	Plastics debris	n.a.	2,400 ± 3,700	
2,4-DTBP	Geoje coast, South Korea	New plastics	n.a.	4,100 ± 6,500	
BHT	Geoje coast, South Korea	Plastics debris	n.a.	1,200 ± 5,400	
BHT	Geoje coast, South Korea	New plastics	n.a.	2,400 ± 9,600	
UV320	Geoje coast, South Korea	Plastics debris	n.a.	2,800 ± 10,000	[14]
UV320	Geoje coast, South Korea	New plastics	n.a.	4,100 ± 15,000	
UV326	Geoje coast, South Korea	Plastics debris	n.a.	4,100 ± 15,000	
UV326	Geoje coast, South Korea	New plastics	n.a.	10,000 ± 36,000	
UV327	Geoje coast, South Korea	Plastics debris	n.a.	1,300 ± 4,100	
UV327	Geoje coast, South Korea	New plastics	n.a.	2,100 ± 7,400	
UV328	Geoje coast, South Korea	Plastics debris	n.a.	110 ± 320	
UV328	Geoje coast, South Korea	New plastics	n.a.	70 ± 160	
Phthalic acid esters (PAEs)	Estuarine mudflats in China	PE pellets	4–5 mm	0.033 ± 0.039	[15]
	“Wild” beach in China	PE pellets	4–5 mm	0.11 ± 0.17	
	Bathing Beach in China	PE pellets	4–5 mm	0.2 ± 0.37	
	Estuarine mudflats in China	PS foams	1–4 mm	1.43 ± 2.33	
	“Wild” beach in China	PS foams	1–4 mm	16 ± 32	
	Bathing Beach in China	PS foams	1–4 mm	4.17 ± 3.22	
	Estuarine mudflats in China	PP flakes	<1 mm	4.34 ± 2.39	
	“Wild” beach in China	PP flakes	<1 mm	15 ± 11	

(continued)

Table 1 (continued)

Chemical additives	Sampling locations	Plastic types	Particle size	Concentration (ng/g)	References
	Bathing Beach in China	PP fragments	1–4 mm	1.73 ± 1.27	
	"Wild" beach in China	PP fragments	1–4 mm	0.83	
	"Wild" beach in China	PE fragments	1–4 mm	27	
Organophosphorus esters (OPEs)	Estuarine mudflats in China	PE pellets	4–5 mm	14 ± 20	[15]
	"Wild" beach in China	PE pellets	4–5 mm	17 ± 30	
	Bathing Beach in China	PE pellets	4–5 mm	70 ± 130	
	Estuarine mudflats in China	PS foams	1–4 mm	17 ± 25	
	"Wild" beach in China	PS foams	1–4 mm	14,100 ± 34,500	
	Bathing Beach in China	PS foams	1–4 mm	970 ± 2,500	
	Estuarine mudflats in China	PP flakes	<1 mm	1,080 ± 1,250	
	"Wild" beach in China	PP flakes	<1 mm	710 ± 1,010	
	Bathing Beach in China	PP flakes	<1 mm	27 ± 18	
	"Wild" beach in China	PP fragments	1–4 mm	69	
	"Wild" beach in China	PE fragments	1–4 mm	120	
	Calblanque in Spanish	PE/PP/PS	<5 mm	385.66 ± 594.52	[16]
	Cape cope in Spanish	PE/PP	>5 mm	205.26 ± 782.72	
	La Llana in Spanish	PA	n.a.	51.39 ± 39.30	

n.d. no detected, *n.a.* data is not available

[a]Non-flame retardants

[b]Flame retardants

antioxidants were present at higher concentrations than UVSs in plastic debris and Irganox 1076 and Irganox 1010 were the most commonly measured antioxidants [12].

An intensive monitoring of HBCD levels was conducted for the EPS debris and microplastics collected from the marine coasts of South Korea and 12 other countries in the Asia-Pacific region. HBCD was detected extensively in EPS buoy debris and EPS microplastics stranded along the Korean coasts; the highest measured concentration was 8,670 μg g^{-1} [7]. Recycled buoys had the highest HBCD levels, followed by microplastics, large buoys, and small buoys. The ɣ-HBCD dominated diastereomeric patterns in the floating buoys as well as in the EPS microplastics. HBCD was also abundantly detected in EPS debris collected from the Asia-Pacific coastal region, with the highest concentration found in Alaskan beach of the USA. This indicated that HBCD contamination via EPS debris was a common environmental issue worldwide.

Recently, Zhang et al. [15] investigated OPEs and PAEs in the beached microplastics collected from North China, and they found that the PP flakes and PS foams contained the highest concentrations of the two additives in contrast to the PE pellets which contained the lowest. The tris(2-chloroethyl)-phosphate (TCEP), tris(1-chloro-2-propyl) phosphate (TCPP), and di(2-ethylhexyl) phthalate (DEHP) were the three predominant compounds found overall. The maximum Σ4 OPE concentration was measured up to 84,595.9 ng g^{-1} in the PS foams. The OPEs were also measured for the plastic debris sampled at three Iberian Peninsula Southeastern beaches [16]. In this study, the OPEs were the most abundant compounds in comparison to PAH, OCPs, and pesticides extracted from the plastic debris.

2.3 Migration and Release of Chemical Additives from the Plastic Debris or Microplastics

The additives can potentially migrate and be released from polymers during the exposure in environments [17]. Therefore, it is of great interest whether organic chemicals from plastic debris or microplastics cause serious environmental risks. Until now, the majority of studies on the migration and fate of plastic additives associated with plastic particles have focused on leaching of flame retardants [16], plasticizers [18], and fluorescent additives [19].

HBCD in Styrofoam buoys massively used in oyster culture farms contributed to enrichment of HBCD in surrounding sediment [20]. Paluselli et al. [18] explored the migration of PAEs from the PE plastic garbage bags and PVC cables as affected by abiotic and biotic factors. This study indicated that light and bacterial exposure increased the total amount of PAEs released from PVC cables by a factor of up to 5, whereas they had no influence in the case of PE bags. In addition to the light and microbes, the water pH, salinity, and organic matter all had impacts on the leaching of chemicals from the plastics. In another study [19], the researcher investigated the

leaching behavior of fluorescent additives from polyurethane sponge microplastics. They found that the additives amount in the water followed the order of basic water > saline water > seawater > West Lake > River >Wetland, which all showed increasing trends with solution pH and leaching time. Tris-2-(chloropropyl) phosphate which has been measured with high concentration in the microplastics was also found having a high desorption ratio from the plastic debris [16].

Nevertheless, release of plastic additives in the intestinal tracts of aquatic species seems to lower than that in environment. Koelmans et al. [21] assessed the potential of leaching of nonylphenol (NP) and bisphenol A (BPA) in the intestinal tracts of *Arenicola marina* (lugworm) and *Gadus morhua* (North Sea cod) by using a biodynamic model. They found that leaching of NP and BPA concentrations was below the lower ends of global NP and BPA concentration ranges. However, the leaching rates of various additives in environmental conditions, as well as organisms' gut conditions, need to be investigated because the amount of various additives in plastic may be very high.

3 Sorption and Desorption of Organic Pollutants in the Microplastics

3.1 Occurrence of Organic Pollutants in the Environmental Microplastics

Studies on the organic pollutants in the environmental microplastics started from monitoring of persistent organic pollutants (POPs) in the plastic resin pellets (small granules 0.1–0.5 cm in diameter). A range of organic micro-pollutants (including polychlorinated biphenyls (PCBs), DDE, and nonylphenol) have been detected in plastic resin pellets stranded on beaches [11]. There is a project named "International Pellet Watch" sponsored by Japanese scientists, which has been in operation since 2006. In the project, the collected plastic resin pellets from beaches around the world were mailed to the laboratory of Japan for POPs analysis, and a total of 30 samples of pellets from 17 countries have been analyzed by 2009 [22]. By using this dataset, the global spatial distributions of organic micro-pollutants could be mapped with extremely low-cost method. Surveys for some specific area have been also carried out at Portuguese coast [23], South Atlantic [24], North China [25], and even at the remote islands in the Pacific, Atlantic, and Indian Oceans [26]. All these surveys focused on the persistent organic pollutants including PAHs, PCBs, DDTs, and HCHs. In addition to the pellets from the surface of the beaches, some authors compared the plastic pellets from the different depths of the sediment of the beaches and found that the concentration and composition of PAHs varied greatly with the depth [27].

Meanwhile, field adsorption experiments using PP virgin pellets demonstrated significant and steady increase in PCB and DDE concentrations throughout the

6-day experiment, indicating that adsorption of PCBs and DDE from the ambient environments by the pellet surfaces is the mechanism of enrichment [11]. Furthermore, a longer term of field measurement of sorption of PCBs and PAHs to five types of plastic pellets demonstrated that sorption rates and concentrations of PCBs and PAHs varied significantly among plastic types and among locations, PE and PP pellets having a higher sorption capacity than PET and PVC [28].

The concentration of organic pollutants in the plastic resin pellets was found having a relationship with the aging time of the pellets in environment. Fifty-five resin pellets from a beach in Tokyo were individually analyzed for PCBs and showed discolored (e.g., yellowing) pellets contained more PCBs than others on most of the beaches sampled [29]. Analogous to the results in Tokyo beach, aged and black pellets were also measured higher concentrations for the PCBs, PAHs, and DDT in the beaches of the Portuguese coast [23]. The increase of adsorption of POPs in the plastic pellets with the aging time may result from the increase of specific surface areas and crystallinity after a long-term exposure in environment [30, 31].

3.2 Sorption and Desorption of Hydrophobic Organic Pollutants in Microplastics

Microplastics were found having a high affinity for hydrophobic organic pollutants (HOCs) due to their high hydrophobicity and specific surface area [32, 33]. Sorption of HOCs by plastic polymers was mainly governed by hydrophobic interaction mechanism. Liu et al. [34] studied the interactions between microplastics and phthalate esters and found that the sorption of DBP was much higher than DEP on all the three tested microplastics due to the higher K_{ow} and lower solubility of DBP than DEP. For the polymers (e.g., PS) with benzene rings in their structure, π-π interaction was supposed to be one of the most important mechanisms for their strong sorption for HOCs. In the same study [34], the higher sorption of DEP and DBP by PS compared to PE could be resulted from the strong π-π interactions between PS and the two PAEs. The previous study also revealed that sorption of PAH to virgin polystyrene microplastics was higher compared to sorption to nonaromatic polyethylene, polyvinyl chloride, and polypropylene microplastics due to the strong π-π interactions between the PS and PAHs [35]. Velzeboer et al. [36] suggested that the strong sorption of PCBs to nano-PS particles in their experiment could be explained by both hydrophobic and π−π interactions. Sorption experiment with nano-PS and PAHs indicated that the adsorption isotherms were nonlinear and a high distribution coefficient up to 10^9 L/kg was obtained as a result of the π−π interactions between the planar PAH and the surface of the aromatic polymer polystyrene [37].

Sorption of HOCs to microplastics might be limited by diffusion in the plastic phase, which could be described as biphasic process, the fast sorption/desorption in the outer layer followed by slow diffusion into the inner plastic phase. This biphasic

model is frequently used for sorption of HOCs to soils or sediments [38]. A recent study showed that the sorption process of HOCs to the micro-sized PS included two stages: the fast sorption stage for the HOCs diffusing through aqueous boundary layer and the slow one for the HOCs penetrating inside the PS particle, while the mass transfer rates were extremely slow for the second sorption phase [39]. The kinetics study on the sorption of pyrene onto microplastics revealed that the sorption rates were mainly controlled by intraparticle diffusion [40]. Furthermore, the penetrating diffusion process was controlled by the molecular weight of HOCs and the polymer density. Fries and Zarfl [41]) studied the sorption of PAHs to low- and high-density PE and found that the diffusion coefficients decreased while the molecular weight of the PAHs increased which indicates a hindered diffusion through the matrix as a result of a larger molecule size. Meanwhile, higher diffusion coefficients were derived for LPDE than for HDPE indicating a greater sorption velocity for LPDE according to the lower polymer density.

Several factors have impacts on the sorption of HOCs onto the microplastics. In terms of the plastic polymers, physical structures have been found to play an important role in the HOC sorption. Guo et al. [32] examined the sorption behavior of four hydrophobic organic contaminants by the different polymers and found that the organic carbon content-normalized sorption coefficients (K_{oc}) of phenanthrene, lindane, and naphthalene by PEs of same composition but distinct physical makeup of domains increased with their crystallinity reduction.

Weathering in environments of the microplastics changes their surface properties and hence alters their sorption behavior for HOCs. The weathered plastics would increase the adsorption capacity than virgin plastics [22, 29]. This might be mainly attributed to the increase in surface area due to polymer weathering that would increase the effective diffusivity and the additional sorbents attached to the plastic debris for hydrophobic contaminants [11]. An increase in the surface area due to weathering could also increase polarity of the polymer surface by introducing oxygen-containing groups, which could decrease the affinity for hydrophobic compounds [29, 42]. Sorption coefficients of naphthalene by polystyrene microplastics following aging were found up to one order of magnitude lower than for pristine particles [43].

External factors, such as temperature, salinity, and composition of the water phase (e.g., particulate and dissolved organic matter), can also influence the sorption behavior of HOCs by microplastics in waters. The temperature has an impact on the sorption of HOCs in the microplastics through changing the surface tension of solution and the solubility of HOCs in the solution. Zhan et al. [44] observed that increasing temperature decreased the sorption of PCB by PP in pure water. The salinity would impact the solubility of hydrophobic organic compounds by natural sorbents such as soils, clays, and sediments [45]. Recent experiment results revealed that the sorption capacity of PCB, phenanthrene, and PAEs in the simulated seawater is higher than those in the ultrapure water, which indicates that salinity acts as one main factor affecting sorption capacity [34, 44, 46, 47]. However, the effects were not the same for different microplastics. In a previous study, it was found that differences in salinity had no significant effects on phenanthrene sorption onto

ultrahigh molecular weight (UHMW) polyethylene microplastics [48]. Dissolved organic matter (DOM) contains abundant functional groups, which can interact with natural particles or organic pollutants and thus affect their fate and transport in the nature environment. In plastic water system, molecular sieving and pore blockage By DOM could have more important roles in the control of the sorption of HOCs by PS than by other polymers [47].

3.3 Sorption and Desorption of Hydrophilic Organic Pollutants in Microplastics

Compounds with more hydrophilic properties have been much less considered in comparison to the highly hydrophobic compounds with respect to their adsorption and desorption in the microplastics. Pharmaceuticals and personal care products (PPCPs), as emerging contaminants (ECs) in terrestrial environments, threaten the aquatic and soil resources. Most of the PPCPs have hydrophilic properties and have a high possibility of interacting with microplastics, especially aged microplastics, because of their hydrophilic, oxygen-containing functional groups [47, 49, 50]. The mechanism governed adsorption of pharmaceuticals in the microplastics included the partitioning, electrostatic interactions, intermolecular hydrogen bonding, and $\pi-\pi$ interactions. A study regarding the adsorption of antibiotics on the different types of microplastics showed that the adsorption capacities of ciprofloxacin (CIP), trimethoprim (TMP), and sulfadiazine (SDZ) on PS are higher than those on PE [51], suggesting the $\pi-\pi$ interactions dominated the adsorption of the three antibiotics at the aromatic surface of the PS. In another study which examined the adsorption of oxytetracycline to microplastic polystyrene, the results revealed that electrostatic interaction regulated the adsorption; meanwhile H-bonding and multivalent cationic bridging mechanisms may also have affected the adsorption [50]. Aging has a pronounced effect on the adsorption enhancement by microplastics owing to the increasing of surface areas, hydrophilic properties of the surface, and oxygen-containing functional groups after aging [49, 50].

Impacts of pH, ionic strength, and dissolved organic on the adsorption have been examined in the present studies. In contrast to the negligible effects of pH on the adsorption of HOCs by microplastics [34], the solution pH has a pronounced effect on the antibiotic adsorption by microplastics because various antibiotics will exhibit different speciation of the cation, zwitterion, and anion in a specific pH condition [49, 50, 52]. For example, the oxytetracycline was mainly in cationic form at $pH < 3.27$, predominated by a zwitterion at pH values 3.27–7.32 and dominated by anions (OTC^{-} and OTC^{2-}) when $pH > 7.32$. A study indicated that the maximal adsorption of oxytetracycline on the aged PS particles occurred at $pH = 5$, because the surface charge of the aged PS approached the point of zero charge (PZC 4.96) at $pH = 5$ and therefore has the maximal adsorption due to the lowest electrostatic repulsion between the oxytetracycline and PS surface [50].

Ionic strength of the solution plays an important role in the regulation of antibiotic adsorption by microplastics. Usually a reduction of adsorption will be observed with the increasing of ionic strength owing to the competing with antibiotics on the adsorption sites on the plastic surface [50–52]. However the ions of different valence state may have different influence on the adsorption. The depression of oxytetracycline sorption to the aged PS foams can be offset in the presence of Ca^{2+} through formation of ternary complexes between the cations and oxytetracycline and the surface functional groups [50].

The effects of DOM on the antibiotic adsorption were not consistent among the different studies. Xu, Liu, Brookes, and Xu [53] found the increasing concentration of fulvic acid inhibited the sorption of tetracycline on three microplastics, decreasing them by more than 90% at the fulvic acid concentration of 20 mg/L. However in the study of Zhang et al. [50], both fulvic acid and humic acid promoted the adsorption of oxytetracycline in the aged PS foams, and humic acid has more pronounced effect than fulvic acid. Such a difference is probably caused by the different microplastics used in the experiments. Further studies are recommended to elucidating the mechanisms of DOM effects on the sorption and desorption of antibiotics in the microplastics.

4 Microplastics Move Additives and Organic Pollutants from Environment to Organisms

4.1 The Role of Microplastics in the Transfer and Accumulation of Chemicals from Environment to Organisms

The vector concept has been used to describe increased uptake of contaminants that adhere to microplastics by planktivores [3]. However, the relative role of microplastics as a vector for hazardous contaminants to organisms has been found negligible in comparison to natural exposure pathways in marine ecosystems [54, 55]. Hartmann et al. [56] still argued that microplastics will play a larger role as a pathway for contaminants to transfer into biota than the current estimation in some specific scenarios. Moreover, microplastics might accumulate in terrestrial and continental food webs at levels similar to or higher than in marine counterparts, although conclusive evidence is yet to be found [1]. Therefore, the process of microplastics as a carrier of contaminants from external environment to biota should not be overlooked in the terrestrial environments.

A study regarding the effects of PS on the transfer of PCBs to lugworms *Arenicola marina* (L.) indicated that a low PS dose of 0.074% increased bioaccumulation of PCBs by a factor of 1.1–3.6, an effect that was significant for ΣPCBs and several individual congeners [57]. Browne et al. [58] also found the vector effect of 5% PVC on transfer pollutants and chemical additives into the gut of

lugworms. Using the ^{14}C-labeled phenanthrene, Ma et al. [59] observed that presence of nanometer plastics significantly enhanced bioaccumulation of phenanthrene-derived residues in daphnid body. The enhanced transport of contaminants by nanoplastics was in relation to the polarity-dependent extents of desorption hysteresis, effective nonpolar and weakly polar compounds, and no effects for polar compounds [60].

Notwithstanding, latest studies showed that pharmaceutical bioaccumulation in the biota could also be enhanced in the presence of microplastics. The PS microplastics were found to enhance the bioaccumulation of roxithromycin in the tissues of freshwater fish red tilapia (*Oreochromis niloticus*) compared to roxithromycin-alone exposure [61]. Moreover, the bioaccumulation factor (BAF) of venlafaxine and O-desmethylvenlafaxine in loach tissue amplified more than ten times with microplastics present, and in liver subcellular structure, microplastic may help to transport more compounds into subtle areas and postpone the contaminant metabolism in organisms [62]. Therefore, these studies contributed to our understanding of the aquatic risks of pharmaceuticals associated with microplastics.

The promotion of contaminant bioaccumulation in the presence of microplastics probably is related with desorption rate of the adsorbed contaminates inside the biota. A laboratory gut mimic extraction using the digestive enzyme pepsin at pH 2 from the commonly-ingested plastics indicated that bisphenol A (BPA), diethylhexyl phthalate (DEHP), and butylbenzyl phthalate (BBP) concentrations were significantly increased in the gut mimic extraction relative to water extraction [63]. Another estimation using artificial gut fluid of fish indicated the microplastic ingestion might increase the total uptake rate of pentachlorobenzene and hexachlorobenzene due to their accelerated desorption from microplastics into the artificial gut solution under the scenario of extremely high intake of microplastics [64]. However, when in comparison with the biochar and wood, the plastic absorbed PCBs had a lower solubilization in the gut fluids [65]. Therefore, the contribution of the organic pollutants to aquatic organisms from microplastics still has a high uncertainty, and further studies are recommended to be carried out in this aspect.

4.2 Effects of Leachates from the Microplastics on the Terrestrial Organisms

One of the ecological risks associated with microplastics contamination in the environment is the release of chemicals associated with plastics to the environment shared with organisms. Most of the information available from this route of exposure comes from experiments assessing the toxicity of leachates from new plastic consumer products to aquatic invertebrates [19, 66–69]. Varied chemicals have been measured in the plastic leachates, which included HOCs [69], dioxin-like compounds [30], and endocrine-disrupting chemicals (EDCs) such as estrogens, bisphenol A, bisphenol S, octylphenol, and nonylphenol [70]. Higher concentrations of these chemicals were measured in the leachates from aged or small-sized

microplastics. In addition, the chemical concentration from the microplastics depends mainly on water environments. Luo et al. [19] found that the leached concentrations of fluorescent additives in simulated and natural water followed the order of basicwater > salinewater > seawater > West Lake > river > wetland.

The toxicity of the leachates was evaluated using the bioassay including the mortality of freshwater species *Daphnia magna* [69, 71], embryo development of the brown mussel (*Perna perna*) and sea urchin *Paracentrotus lividus* [66, 68], larval survival and settlement of the barnacle (*Amphibalanus Amphitrite*) [67], and microalgae (*Chlorella vulgaris*) physiology [19]. All these tests were observed toxic effects of the plastic leachates. Moreover, the toxicity of the leachate from beached pellets was found much higher than that of virgin pellets, which corresponds to the higher concentrations of the contaminants in the aged microplastics [30, 66, 68]. Alteration of the surface chemistry of the microplastics may also have an important impact on the toxic effects. Study of Li et al. [67] revealed that hydrophobicity of the plastic surface was negatively correlated with mortality of barnacle larval when it was tested in the plastic leachates for a short time. This indicated that increasing of hydrophilicity on the plastic surface as a result of weathering may enhance the toxic of the microplastics. However, longer-term studies are required to determine if relationships persist as plastics become weathered by environmental exposure.

4.3 Biological Consequences of Microplastic Ingestion and Chemical Transfer to Organisms

We know that microplastics are easy to be swallowed and accumulated by aquatic organisms [3] and, consequently, be transferred through food chains [72]. Therefore, the importance of combined ecological effects caused by microplastics and organic pollutants has been emphasized since microplastics can act as a vector for most of the organic pollutants [33]. The endpoint of the risk assessment involved mortality, feeding behavior, immunity, and biomarkers for oxidative stress in the bioassays. However, contrasting results have been obtained based on current literature review. A short-term experiment with large proportions of PVC (5%) showed that lugworms eating microplastics accumulated large enough concentrations of pollutants (triclosan) or additives (PBDE, nonylphenol) which enhanced the reduction of survival, feeding, and immunity of the lugworms [58]. Another study indicated that the co-effect of microplastics and venlafaxine, as well as the metabolite, might lead to more adverse effect against loach and therefore should be taken into consideration in actual environment [62].

However, more studies indicated that the toxicity of organic pollutants on the biota was not affected or even mitigated in the presence of microplastics. For example, the microplastics delayed pyrene-induced goby (*Pomatoschistus microps*) mortality and increased the concentration of bile pyrene metabolites, and simultaneous exposure to both microplastics and pyrene did not increase significantly the inhibitory effect

for acetylcholinesterase (AChE) and isocitrate dehydrogenase (IDH) activities [73]. Co-exposures and incubated exposures of microplastics and fluoranthene did not result in additive or synergistic oxidative stress response in the blue mussel, *Mytilus edulis* [74]. The combination of triclosan and PVC microplastics even has a greater reduction of their toxicity on microalgae *Skeletonema costatum* than the microplastics alone [75]. Although a significant inhibition in the activity of 7-benzyloxy-4-trifluoromethyl-coumarin O-dibenzyloxylase (BFCOD) enzyme was observed in the co-exposure to roxithromycin (ROX) and PS microplastics compared to exposure to ROX alone, increase of SOD activity and decline of MDA content caused by co-exposure suggested that presence of microplastics might mitigate the oxidative damage [61].

In addition to the bioassay using the spiked organic contaminants with microplastics, Asmonaite et al. [76] applied the sewage or harbor effluent exposed PS microplastics which contained various environmental contaminants (e.g., PAHs, nonylphenol and alcohol ethoxylates, and others) to feed fish rainbow trout and examined the hepatic stress and lipid peroxidation in fish fillet. The results indicated that the ingestion of relatively high doses of these PS microplastics did not induce adverse hepatic stress in fish liver and the ingestion of these particles did not affect lipid peroxidation or rancid odor development, thus not affecting fillet's quality.

5 Summary

Environmental microplastics contain a variety of contaminants involved of adsorbed organic pollutants and chemical additives. Hence both microplastics and the contained organic pollutants should be considered with respect to the ecological risk caused by the microplastics. The microplastic-bound contaminants have different adsorption and desorption characterizations depending on the characteristics of the contaminants, physicochemical properties of the microplastic surface, and external environmental conditions. Aging of the microplastics in the environment would have important impacts on its surface properties and subsequently influence the adsorption for organic pollutants. Therefore, further studies are recommended to study on the interaction of the organic contaminants with environmental relevant microplastics, and fate of these contaminants in the environment. There are a lot of uncertainties regarding the organic contaminant bioaccumulation and toxic effects owing to the co-exposure of microplastics and organic pollutants to the biota. Meanwhile, researchers are concerned more about the polymer types of the microplastics than the size and shape difference in most of the studies regarding the combined effects of microplastics and organic pollutants. Hence the role of physical properties of the microplastics should also be of concern in the bioassay of co-exposure of microplastics and organic pollutants in order to provide an unbiased evaluation.

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